

Response Surface Methodology-Driven Optimization of Flocculation-Assisted Wastewater Remediation Using Tamarind Seeds

Katihalli Lakshme Gowda Yashwanth¹ and Pallavi Nagaraju^{1*}

¹Department of Environmental Sciences, JSS Academy of Higher Education & Research, Mysuru.

ABSTRACT

Freshwater scarcity, representing barely 1% of the global water reserve, constitutes a critical challenge to human health, environmental sustainability, and socio-economic development. Currently, nearly 40% of the world's population resides in arid and semi-arid regions, a figure projected to exceed two-thirds by 2025. Concurrently, rapid industrialization, escalating pollution loads, and progressive environmental degradation have intensified pressure on limited water resources. In this context, the exploration of sustainable and naturally derived treatment materials has gained substantial importance.

The present investigation focuses on the application of a naturally occurring bio-mineral adsorbent derived from tamarind seeds for wastewater remediation. Response Surface Methodology (RSM), a robust statistical and mathematical optimization tool, was employed to model and evaluate the interactive effects of critical operational parameters influencing pollutant removal efficiency. Owing to their high surface area, porous structure, and abundant functional groups, tamarind seed-based natural adsorbents exhibit significant potential for contaminant adsorption.

Flocculation-assisted treatment experiments were conducted using tamarind seed dosages ranging from 0.1 to 1.0 g/L under varying pH and contact time conditions. Optimal removal efficiencies were achieved at pH 6.25 with a dosage of 0.1 g/L, yielding 74% turbidity reduction; at pH 6.5 with 0.6 g/L dosage, resulting in 41% Chemical Oxygen Demand (COD) removal; and at pH 6.5 with 1.1 g/L dosage, achieving 76% Total Solids (TS) reduction over an optimized contact time of 46.4 minutes.

These findings demonstrate that tamarind seed-based bio-adsorbents provide an efficient, low-cost, and eco-friendly alternative for wastewater treatment. The utilization of such organic materials presents a viable strategy for enhancing water quality, enabling safe water reuse, and mitigating associated public health risks while promoting sustainable environmental management.

Keywords: Tamarind seed, turbidity, bio-coagulant, coagulation-flocculation, response surface methodology.

1. INTRODUCTION

Water is indispensable for sustaining life and socio-economic development. The availability of adequate quantities of safe and potable freshwater has historically governed the establishment and expansion of human civilizations, particularly around rivers and lakes

that supported transportation, agriculture, and domestic consumption. Early assessments of water quality relied primarily on sensory perception, including taste, odour, and visual clarity, prior to the advancement of biological, chemical, and medical sciences that enabled systematic evaluation of water safety and its implications for public health (Shoba et al., 2015).

Rapid industrialization, urbanization, and population growth have intensified the discharge of untreated effluents into aquatic systems, resulting in severe deterioration of water quality. Anthropogenic inputs such as municipal sewage, domestic runoff, industrial effluents, mining residues, and agricultural leachates enriched with fertilizers and pesticides constitute major sources of contamination (IJSER172323, n.d.). Wastewater typically contains a complex mixture of suspended and dissolved pollutants, imparting turbidity, odour, and toxicity, thereby rendering it unsuitable for potable and domestic use (Amran et al., 2021; Natarajan et al., 2018). Municipal wastewater, in particular, comprises water from residential, commercial, and institutional activities mixed with organic wastes including urine and faecal matter (Joaquin et al., 2021).

Among conventional treatment technologies, coagulation–flocculation remains one of the most widely adopted physicochemical processes for the removal of colloidal and suspended particles from wastewater. Coagulation destabilizes charged particles to form gelatinous precipitates, while flocculation promotes gentle aggregation through controlled mixing to generate settleable flocs (Amran et al., 2022; Onukwuli & Okey-Onyesolu F, 2015). Process efficiency depends on operational parameters such as coagulant type and dosage, pH, mixing intensity, and wastewater characteristics (J et al., 2017; Joaquin et al., 2021).

However, the extensive use of chemical coagulants—particularly aluminium and iron salts such as $\text{Al}_2(\text{SO}_4)_3$, $\text{Fe}_2(\text{SO}_4)_3$, AlCl_3 , and FeCl_3 —poses economic and environmental concerns (Nimesha et al., n.d.; Sujana et al., 2019). Alum, one of the most commonly used coagulants, reacts with bicarbonate alkalinity to form gelatinous precipitates that facilitate suspended solids removal (Singh & Kishor, n.d.). Despite its effectiveness, residual aluminium in treated water has been associated with potential health risks, including neurological disorders, and may significantly alter treated water pH while increasing sludge generation and treatment costs (Zainol & Nasuha Mohd Fadli, 2020; Nair et al., 2008).

These limitations have driven increasing interest in natural, plant-based coagulants as sustainable alternatives. Natural coagulants are biodegradable, renewable, non-toxic, cost-effective, and locally available (Ayangunna et al., n.d.; Thesni et al., 2021). Tamarind (*Tamarindus indica*) seed powder has emerged as a promising bio-coagulant owing to its polysaccharide and protein content, which facilitate particle aggregation. It is edible, eco-friendly, and capable of reducing key pollution indicators such as turbidity, chemical oxygen demand (COD), and total solids (TS) (Koul et al., 2022; R et al., n.d.). Extensive research has therefore focused on modifying and optimizing plant-derived coagulants for enhanced treatment performance (Sivakumar et al., 2014; Kaushal & Goyal, n.d.; Badawi et al., 2023; Ndabigengesere & Narasiah, 1998).

To systematically optimize treatment efficiency, statistical and mathematical modeling tools such as Response Surface Methodology (RSM) have gained prominence. RSM enables evaluation of individual and interactive effects of operational variables through reduced experimental trials while generating predictive quadratic models (Kumar et al., 2022; Joaquin et al., 2021). The methodology involves experimental design selection, variable optimization, model development, graphical response analysis, and identification of optimal operating domains. Owing to its robustness, RSM has been widely applied in wastewater treatment, adsorption studies, and environmental process optimization.

In this context, the present study aims to develop an RSM-based predictive model for the removal of turbidity, COD, and total solids using tamarind seed powder as a natural coagulant. Key independent variables include pH, contact time, and coagulant dosage. Experimental pH adjustments were performed using NaOH and HCl within the design limits generated by JMP Pro 17. The treatment protocol involved rapid mixing at 100 rpm for 3 minutes followed by slow mixing at 40 rpm for 18 minutes and subsequent settling for 20–60 minutes prior to filtration. A total of 13 experimental runs were conducted to evaluate process interactions and optimize pollutant removal efficiency.

2. MATERIALS AND METHODOLOGY:

2.1 Collection of Wastewater Sample

Sewage wastewater samples used in this study were collected from a residential drainage outlet located in Mogarahalli, KRS Road, Mysuru District, Karnataka, India. Samples were obtained in pre-cleaned, high-density polyethylene (HDPE) containers and transported to the laboratory under refrigerated conditions (4 °C) to minimize physicochemical and biological alterations prior to analysis and treatment experiments.

2.2 Preparation of Tamarind Seed Powder

Tamarind (*Tamarindus indica*) seed kernels, an agricultural by-product commonly discarded as waste, were utilized as a natural coagulant in this investigation. Tamarind kernel powder has demonstrated significant efficiency as a bio-clarifying agent for municipal and industrial wastewater treatment.

Conventional treatment systems widely employ aluminium-based coagulants such as alum; however, residual aluminium ions in treated water have been associated with potential health risks, including neurotoxicity and Alzheimer's disease (Meghana & Keshava Kumar, 2020). In contrast, plant-derived coagulants are biodegradable, non-toxic, and environmentally sustainable.

Raw tamarind seeds were procured from local sources and manually decorticated to separate the kernels. The kernels were thoroughly washed with distilled water to remove adhering impurities and subsequently sun-dried for five days to eliminate residual moisture. The dried kernels were then pulverized using a mechanical grinder, and the resulting powder was sieved through a 500-µm mesh to obtain uniform particle size suitable for coagulation–flocculation experiments as depicted in figure 1. (Shoba et al., 2015; Thesni et al., 2021).



Fig 1: (a) Tamarind Seed and (b) Powder.

2.3 Experimental Design and Coagulation–Flocculation Procedure

Coagulation–flocculation is one of the most widely employed physicochemical processes in sewage treatment plants due to its effectiveness in removing suspended and

colloidal impurities. In the present study, coagulation–flocculation experiments were conducted using a standard jar test apparatus to evaluate and optimize treatment performance.

The jar test unit consisted of six paddles operated by an independent variable-speed motor, with rotational speeds adjustable from 0 to 150 rpm. Prior to experimentation, collected wastewater samples were thoroughly agitated to ensure homogeneity and to resuspend any settled particulates. Equal aliquots of the sample were then transferred into 1-L beakers for treatment trials. The independent variables selected for the experimental design were: Coagulant dosage ((Tamarind seed powder) 0.1 – 1.1 g/L); pH(4.0 – 9.0); Settling time(40 – 60 min (for optimization in DoE). The pH of wastewater samples was adjusted to the desired range using 1 N H₂SO₄ or 1 N NaOH solutions.

The coagulation–flocculation process was carried out in three sequential stages: Rapid mixing (100 rpm for 3 minutes); Slow mixing ((flocculation): 40 rpm for 18 minutes); Settling (40–60 minutes at 30 ± 2 °C). Following sedimentation, the dense flocculated sludge was carefully separated, and the clarified supernatant was collected for further physicochemical and microbiological analyses. All experiments were performed in quadruplicate, and the mean values of the supernatant characteristics were considered for data interpretation. Treatment efficiency was evaluated based on the following dependent variables: Turbidity, Chemical Oxygen Demand (COD), Total Solids (TS), Bacterial load (CFU)

2.4 Morphological Characterization of Bio-Coagulant

Scanning Electron Microscopy (SEM) analysis was employed to examine the surface morphology of tamarind seed powder. SEM is a high-resolution imaging technique widely used in environmental material characterization for assessing surface texture, porosity, and adsorption sites. For analysis, powdered tamarind kernels were sieved through a 500-μm mesh to ensure uniform particle size. The prepared samples were mounted on SEM stubs, sputter-coated (if required), and examined to evaluate structural features relevant to coagulation and adsorption performance.

2.5 Statistical Optimization Using Response Surface Methodology

Process optimization was carried out using Response Surface Methodology (RSM). Independent variables—coagulant dosage, pH, and settling time—were systematically varied to evaluate their individual and interactive effects on pollutant removal. A custom Design of Experiments (DoE) matrix was generated using JMP Pro software (Version 17). A total of 13 experimental runs were performed as per the RSM design. Quadratic polynomial equations were developed to predict response variables (turbidity, COD, and TS removal efficiencies) as shown in table 1. The statistical model enables prediction of treatment performance, identification of significant variable interactions, determination of optimal operating conditions. Design-Expert/JMP-based graphical outputs, including response surface plots and contour diagrams, were used to visualize process optimization.

Table 1: Custom design trails-RSM method-JMP Pro 17 for tamarind seed.

Sl. No	pH	Coagulant Dosage g/L	Settling time (min)
1	5.25	0.655	60
2	5.25	0.655	60
3	9	0.6	40

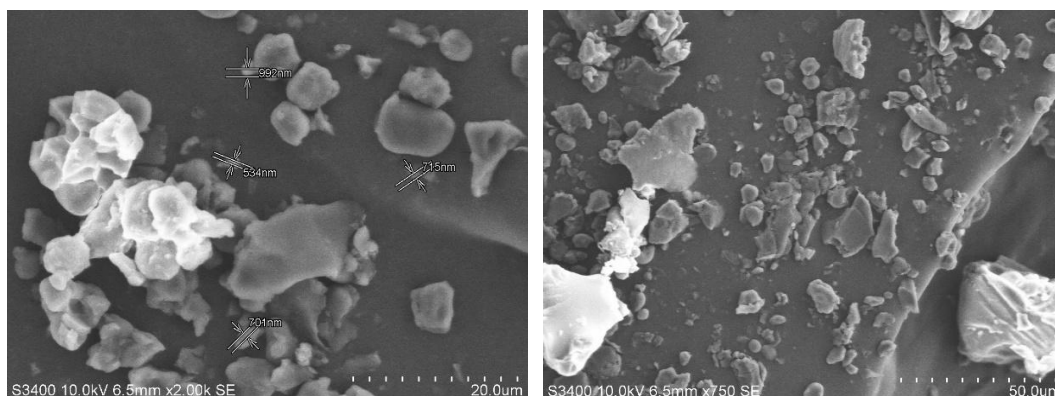
4	9	0.1	60
5	9	1.1	60
6	4	0.6	40
7	4	1.1	50
8	6.25	0.1	40
9	4	0.1	54.4
10	9	0.6	50
11	6.5	1.1	40
12	6.5	0.6	50
13	6.5	0.6	50

3 RESULTS AND DISCUSSION

3.1 Scanning Electron Microscopy (SEM) Analysis

Electron Microscopy (SEM) was employed to elucidate the surface morphology and microstructural characteristics of tamarind (*Tamarindus indica*) seed powder used as a natural coagulant. SEM is a high-resolution surface imaging technique that utilizes a focused electron beam to generate detailed topographical and morphological information, enabling the identification of adsorption-relevant structural features. The FESEM micrograph obtained at a magnification of 2000× (Figure 2) revealed a highly heterogeneous surface architecture. The tamarind seed powder exhibited irregular particle geometry with rough surface texture, fissures, and well-developed porous cavities. Such morphological heterogeneity is advantageous for coagulation–flocculation applications, as it enhances the availability of active binding sites and facilitates particle entrapment.

The presence of surface pores and void spaces indicates a high probability of adsorption and inter-particle bridging mechanisms during treatment. These cavities serve as potential loci for the attachment of suspended particulates, colloids, and organic matter present in wastewater. Furthermore, the rough and uneven surface topology promotes electrostatic interactions and polymeric bridging between the bio-coagulant matrix and destabilized pollutants. The observed microstructural features corroborate the effective pollutant removal efficiencies recorded during jar test experiments. Similar porous and fibrous morphologies reported for plant-based coagulants have been directly associated with enhanced turbidity and organic load removal due to increased surface area and adsorption potential. Overall, SEM analysis confirms that tamarind seed powder possesses favourable morphological characteristics—such as porosity, surface roughness, and structural irregularities—that significantly contribute to its performance as an eco-friendly natural coagulant in wastewater treatment systems.



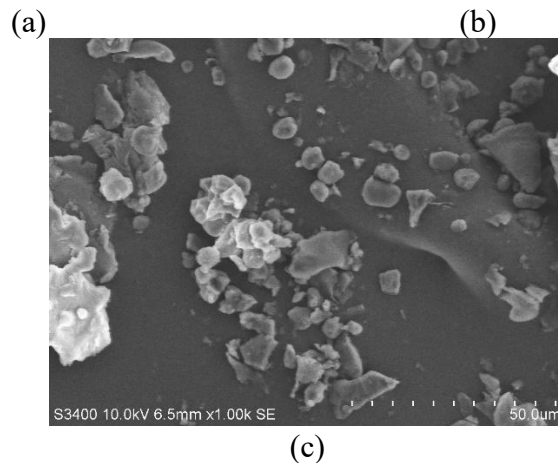


Fig 2 (a) (b) and (c): FESEM microphotography of tamarind seeds powder under 2000x magnification.

3.2 Response Surface Methodology (RSM) Optimization

Pollutant removal performance of tamarind seed bio-coagulant was optimized using Response Surface Methodology (RSM) incorporating three independent variables—coagulant dosage, pH, and settling time—through a 13-run experimental design matrix. The experimental removal efficiencies for turbidity, COD, and total solids are presented in Table 2. RSM modelling enabled evaluation of both individual and interaction effects of process variables, facilitating prediction of optimal operating conditions for maximum treatment efficiency.

Table 2: Final Value of the Municipal Wastewater after Testing of Pollutant Removal Efficiency.

Sl. No	pH	Coagulant Dosage (g/L)	Settling Time (min)	Turbidity removal efficiency (%)	COD removal efficiency (%)	Total Solids removal efficiency (%)
1	5.25	0.655	60	32	16	70
2	5.25	0.655	60	31	33	56
3	9	0.6	40	43	25	60
4	9	0.1	60	55	16	71
5	9	1.1	60	42	16	76
6	4	0.6	40	51	16	76
7	4	1.1	50	55	16	54
8	6.25	0.1	40	74	33	66
9	4	0.1	54.4	45	33	63
10	9	0.6	50	52	16	60
11	6.5	1.1	40	43	33	76
12	6.5	0.6	50	57	33	60

13	6.5	0.6	50	57	41	67
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3.2.1 Optimization of Turbidity Removal

Among the experimental trials, Trial 8 demonstrated optimum turbidity reduction. The ideal operational conditions were: Coagulant dosage: 0.1 g/L; pH: 6.5–7.5; Settling time: ~40 min; Residual turbidity: 6 NTU; Removal efficiency: 74%. The high turbidity removal at relatively low dosage indicates effective charge neutralization and inter-particle bridging by tamarind seed polymers.

Model Adequacy and Residual Analysis - Figure 3 presents the predicted vs. actual turbidity removal efficiency. The regression model exhibited excellent agreement between experimental and predicted values, with a coefficient of determination: $R^2 = 0.99905$. Residual diagnostic plots (Figure 4a & 4b) further confirmed model reliability: Residuals were randomly distributed around zero. No systematic deviation was observed. Studentized residuals fell within acceptable limits. No outliers were detected. This validates homoscedasticity and normal distribution assumptions of the regression model.

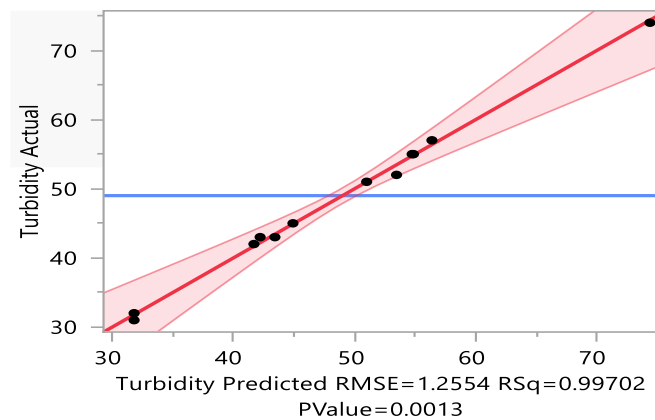
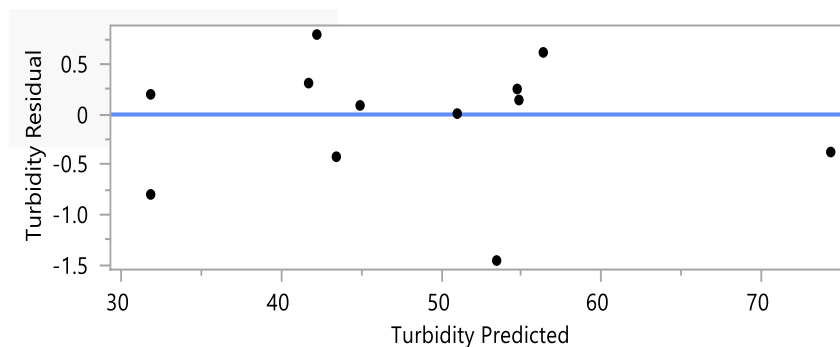
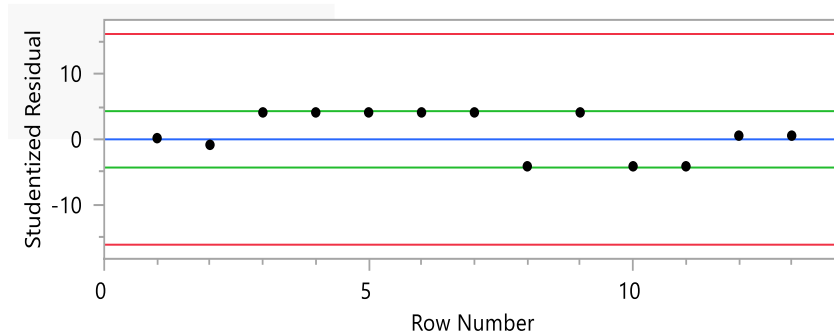


Fig 3. Predicted Vs Actual Removal Efficiency of Turbidity by Tamarind Seed



(a)



(b)

Fig 4(a)&(b): Residual versus predicted graph Studentized residual versus row number.

3.2.2 ANOVA Interpretation for Turbidity Model

Table 3 summarizes the Analysis of Variance (ANOVA). Key statistical indicators include F-ratio: 0.7011; p-value: 0.4905; Max R^2 : 0.8338. Although the lack-of-fit was statistically non-significant, the high regression coefficient and strong predicted–actual agreement confirms model adequacy for process prediction. The high coefficient of determination ($R^2 \approx 0.99$) indicates minimal deviation between experimental and modelled responses, confirming robustness of the quadratic model.

Table 3: Analysis of variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	1	61.87620	61.8762	0.7011
Pure Error	2	176.50000	88.2500	Prob > F
Total Error	3	238.37620		0.4905
				Max RSq
				0.8338

Using an ANOVA-based analysis, the generated model's predictions were verified to be reliable. Important statistical indicators from the ANOVA are provided by the F ratio, p-value, degrees of freedom (DF), sum of squares, mean square, and F ratio; Table 3 details the outcomes. The mean square values are obtained by dividing the total squares of all the components by the error variance and the proper degrees of freedom for the model. The model is then evaluated to determine its relevance using the F-value and p-value. A model is deemed statistically significant if it has a high F-value (0.7011) and a low p-value (0.4905). The coefficient of determination (R^2 square) provides more evidence for the model's fit correctness. In Figure 5, the study's coefficient of determination is calculated as $R^2 = 0.99$. Furthermore, the substantial nature of the generated model is suggested by the high coefficient of determination ($R^2 = 0.99$). The model is appropriate for this investigation because the regression coefficient is near one, meaning that there aren't many disparities between the experimental and anticipated results.

3.2.3 Parameter Significance (Log Worth Analysis)

The relative significance of model terms is illustrated in Figure 5 using Log Worth values ($-\log_{10} p\text{-value}$). A Log Worth value > 2 ($p < 0.01$) indicates statistical significance. Settling time exerted the greatest influence on pollutant removal, followed by dosage and interaction effects. This highlights the importance of floc maturation and sedimentation kinetics in bio-coagulation systems.

Source	Logworth		PValue
Settling Time(40,60)	3.307		0.00049
Settling Time*Settling Time	3.222		0.00060
Catalyst Dosage(0.1,1.1)	3.188		0.00065
Catalyst Dosage*Settling Time	2.853		0.00140
pH*Settling Time	2.709		0.00195
pH*Catalyst Dosage	2.624		0.00237
Catalyst Dosage*Catalyst Dosage	2.582		0.00262
pH*pH	2.150		0.00708
pH (4,9)	1.804		0.01572 ^

Fig 5. Logworth and p-value of the respective parameter and their quadratic components.

3.3 Optimization of COD Reduction

Trial 13 yielded optimal COD removal under the following conditions: Dosage: 0.6 g/L; pH: 6.5; Settling time: ~50 min; Final COD: 700 mg/L; Removal efficiency: 41%. Although coagulation–flocculation is not primarily designed for dissolved organic removal, significant COD reduction was achieved via adsorption and enmeshment within floc matrices.

Model Performance - R^2 : ~0.999; RMSE: 8.914; p-value: 0.5077. Predicted vs. actual plots (Figure 6) demonstrated strong correlation, confirming predictive reliability. Residual diagnostics (Figure 7a & 7b): Residuals aligned along a straight trend; No influential outliers detected; Studentized residuals within Bonferroni limits; This confirms model suitability for COD response prediction.

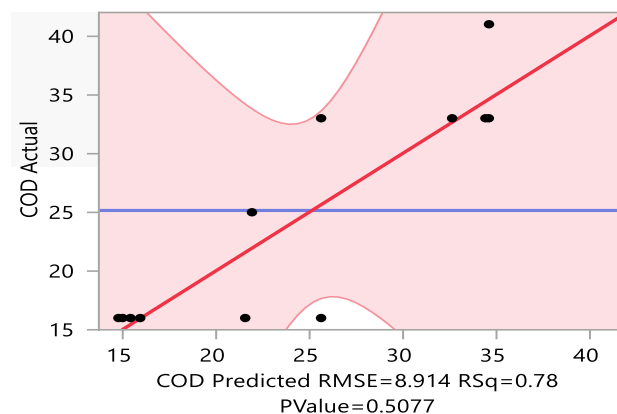
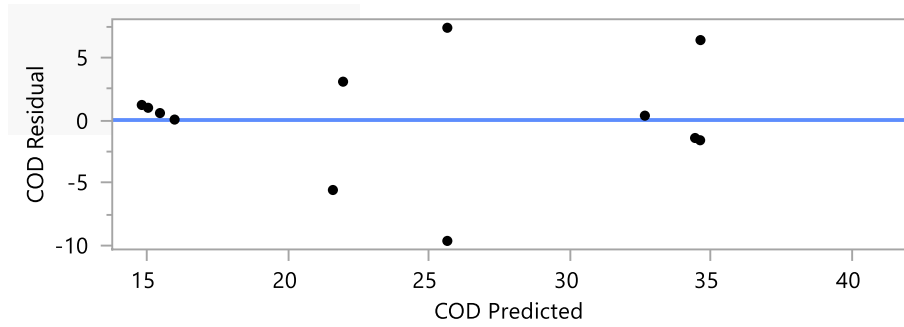
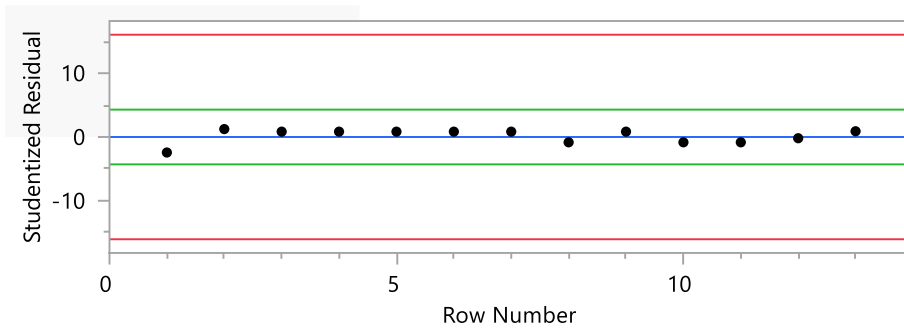


Fig 6. Predicted vs. actual removal efficiency of COD by tamarind seed.



(a)



(b)

Fig 7(a)&(b): Residual versus predicted graph Studentized residual versus row number.

3.3.1 ANOVA for COD Model

Statistical indicators: F-ratio: 0.7011; p-value: 0.4905; R^2 : 0.78. The moderate R^2 suggests acceptable but comparatively lower predictive strength than turbidity, reflecting the limited capability of coagulation alone in removing soluble organics.

Table 4: Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	1	61.87620	61.8762	0.7011
Pure Error	2	176.50000	88.2500	Prob > F
Total Error	3	238.37620		0.4905
				Max RSq
				0.8338

3.4 Optimization of Total Solids (TS) Removal

Optimal TS removal was achieved in Trial 5 under the following conditions: Dosage: 1.1 g/L; pH: 9; Settling time: 60 min; Final TS: 452 mg/L; Removal efficiency: 76%. Higher dosage enhanced sweep flocculation and particle entrapment, leading to superior solids removal.

Model Statistics: R^2 : ~0.999; RMSE: 6.3954; p-value: 0.3891. Predicted vs. actual plots (Figure 8) confirmed strong model agreement. Residual plots (Figure 9a & 9b): Random distribution; No heteroscedasticity; No statistical anomalies

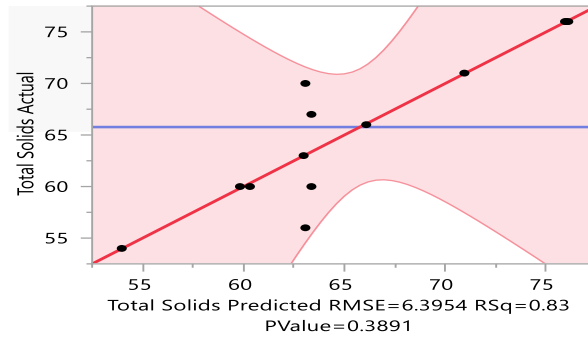
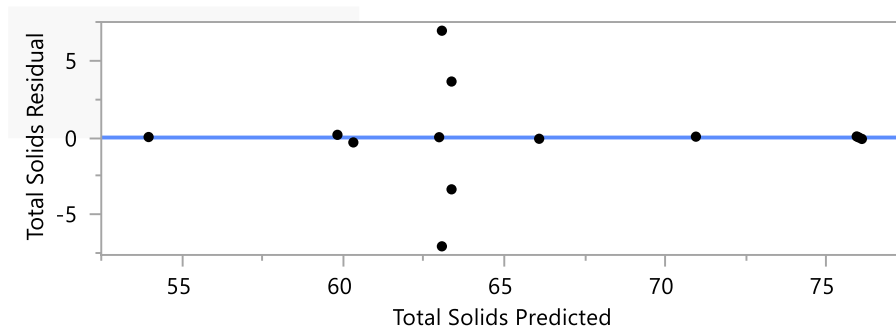
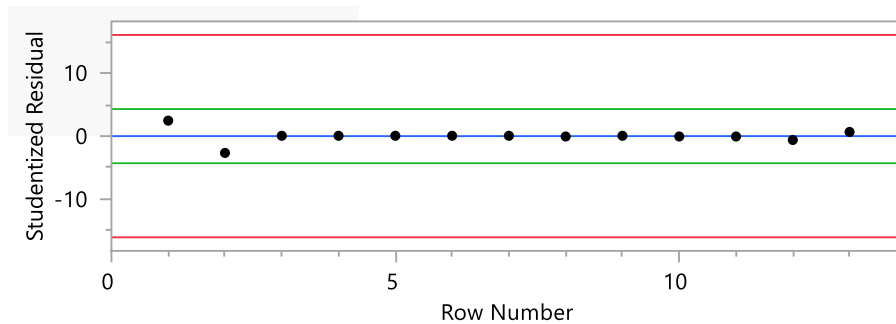


Fig 8. Predicted vs. actual Removal Efficiency of Total Solids by using tamarind seed



(a)



(b)

FIG 9(a)&(b): Residual versus predicted graph Studentized residual versus row number.

3.4.1 ANOVA for TS Model

Table 5: Analysis of variance for total solids. The high R^2 indicates acceptable predictive capability despite low F-ratio, confirming suitability of the quadratic model for TS optimization.

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	1	0.20174	0.2017	0.0033
Pure Error	2	122.50000	61.2500	Prob > F
Total Error	3	122.70174		0.9595

Source	DF	Sum of Squares	Mean Square	F Ratio
				Max RSq
				0.8256

3.5 Three-Dimensional Response Surface Analysis

Three-dimensional response surface and contour plots (Figure 9a–c) illustrate interaction effects of dosage, pH, and settling time on pollutant removal. RSM optimization confirmed that tamarind seed bio-coagulant is highly effective for turbidity and solids removal, with moderate COD reduction capability. Strong regression coefficients, non-significant residual deviations, and well-distributed diagnostic plots validate the statistical robustness of the developed models.

3.5.1 Turbidity Response Surface

Optimal region identified at: Dosage: 0.1 g/L; pH: 6.5; Settling time: 40 min; Removal: 74%. The response surface confirmed strong sensitivity of turbidity removal to dosage–pH interaction.

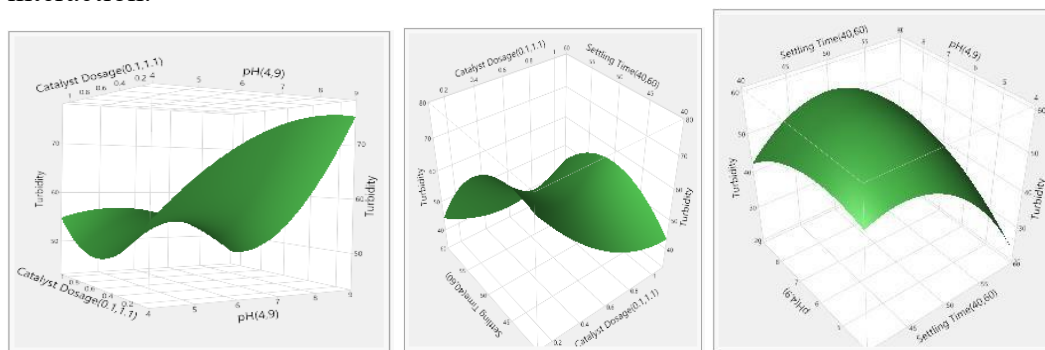


Fig 9a . Three-dimensional response surface plots demonstrate the impact of coagulant dosage and pH on the reduction of Turbidity.

3.5.2 COD Response Surface

Optimal condition: Dosage: 0.6 g/L; pH: 6.5; Settling time: 50 min; Removal: 41%. Surface plots demonstrated moderate interaction between pH and dosage, reflecting partial organic adsorption.

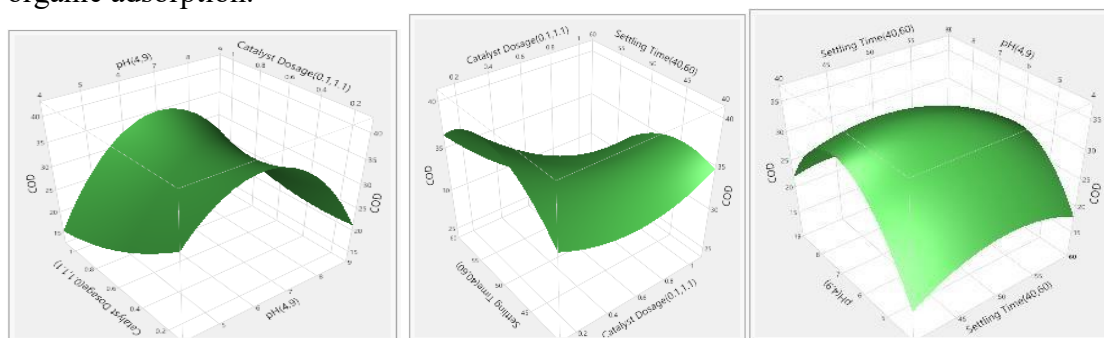


Fig 9b. Three-dimensional response surface plots demonstrate the impact of coagulant dosage and pH on the reduction of COD.

3.5.3 Total Solids Response Surface

Optimal condition: Dosage: 1.1 g/L; pH: 9; Settling time: 60 min; Removal: 76%. Higher alkalinity and dosage enhanced sweep floc formation and sedimentation efficiency.

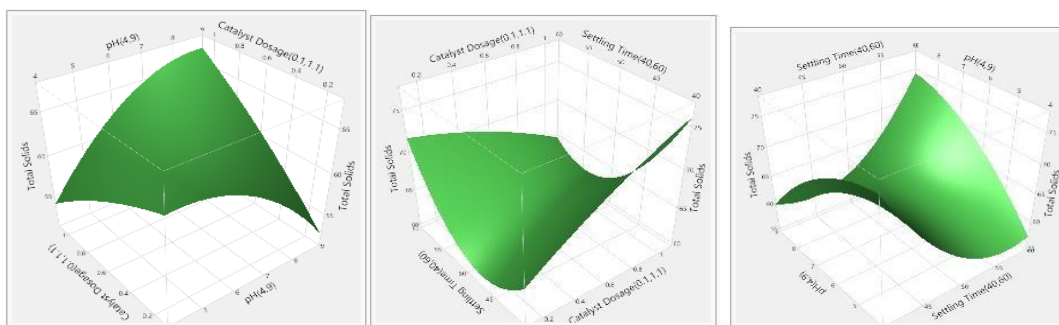


Fig 9c. Three-dimensional response surface plots demonstrating the impact of coagulant dosage and pH on the reduction of Total Solids

CONCLUSION

The present investigation systematically evaluated the efficacy of a natural biosorbent tamarind (*Tamarindus indica*) seed powder for the removal of physicochemical pollutants from domestic wastewater using statistical optimization through JMP software. The biosorbent demonstrated substantial adsorption potential toward contaminant ions, indicating its suitability as a sustainable alternative to conventional chemical coagulants.

Experimental outcomes revealed that tamarind seed exhibited effective pollutant removal across a broad pH spectrum (4–9), with optimal performance recorded under near-neutral conditions (pH 6.5). At this pH, enhanced destabilization of colloidal particles and improved adsorption of dissolved constituents were observed. Process variables—including temperature, mixing duration, solution pH, and biosorbent dosage—were found to significantly influence treatment efficiency, underscoring the importance of multivariate optimization in coagulation–flocculation systems.

Overall, the findings highlight tamarind seed as an economical, biodegradable, and efficient biosorbent for wastewater remediation. Statistical optimization using JMP further strengthens its applicability by enabling precise prediction and control of treatment performance under varying operational conditions. Such optimization enhances scalability and operational feasibility for real-world wastewater treatment applications.

From an environmental sustainability perspective, the adoption of tamarind seed-based treatment offers multiple advantages, including eco-friendly water purification, valorisation of agro-waste, reduced chemical dependency, and cost-effective process integration. Broader implementation could support environmental restoration, safeguard public health, and improve community welfare, particularly in resource-limited settings.

Importantly, this bio-based treatment strategy aligns with global Sustainable Development Goals (SDGs)—notably SDG-6 (Clean Water and Sanitation) and SDG-3 (Good Health and Well-being)—by promoting sustainable water quality management while mitigating environmental pollution.

DECLARATION

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Author contributions: Katihalli Lakshme Gowda Yashwanth executed the wastewater treatment, analyzed the obtained results, and prepared the first draft of the manuscript. Nagaraju Pallavi assisted in the interpretation of the results and validated the methodology for hypothesized the study.

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