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Research article

Blended natural and synthetic coagulants for the COD and BOD removal from surface water; optimization by response surface methodology: The case of Gibe river

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ABSTRACT

A novel wastewater treatment method is presented in this study. It combines natural coagulants derived from watermelon seeds with the commonly used synthetic coagulant alum. This research demonstrates a remarkable synergy between these two coagulants in removing nutrients from Gibe River wastewater. Combining natural and chemical coagulants often improves water treatment by enhancing particle destabilization, accelerating floc formation, and broadening the range of removable contaminants, resulting in lower chemical dosage requirements. The optimal mixing ratio, found to be 1 part watermelon seed coagulant to 3 parts alum, leads to improved treatment efficiency. At this ratio, the process achieves impressive removal rates: 98.26 % for total dissolved solids (TDS), 96.10 % for biochemical oxygen demand (BOD), and 95.26 % for chemical oxygen demand (COD). These findings not only validate the use of watermelon seeds as a coagulant but also highlight the combined approach's environmental and economic benefits. This integrated method offers a more sustainable and cost-effective solution for wastewater treatment.

1. Introduction

The Gibe River serves as Jimma Town's drinking water source. Different tributary rivers, streams, springs, and other water bodies join the Gibe River at different cross-sections. This water body contains a huge number of contaminants generated in urban and rural areas and makes a single flow to join Jimma's town water treatment plant facility. The constant addition of waste to this river is intensifying, thereby increasing the burden on the treatment process. The cost of treatment work is also rapidly increasing. In recent years, water treatment plants have changed the mindset of water operators, causing them to adopt and execute sustainable development in their operations. Water is an essential requirement for all living things, including humans. Water is one of the most important components of life and is required for survival [1]. Human beings use surface water for a variety of purposes. Surface water includes water from dams, streams, and rivers. Due to its high solubilizing power, surface water contains several tiny particles and impurities dissolved in it [2,3]. Minerals and organic and inorganic compounds are examples of dissolved impurities that alter the

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water's physical, chemical, and biological properties [4]. Animal and human waste, as well as runoff chemicals, may contaminate these sources more easily. It is recommended to treat surface water before drinking it [5]. Therefore, surface water must undergo treatment to meet the expected standards for domestic and agricultural applications. Methods like coagulation, sedimentation, filtration, aeration, and disinfection, are common unit processes for water treatment [6,7]. The type and extent of water contamination, as well as the planned use of treated water, influence the choice of water purification techniques. People frequently use coagulation in the presence of colloids that cause turbidity and non-settling suspensions [8]. One of the most straightforward and cost-effective methods for treating surface water is coagulation [9]. Coagulation is a process that destabilizes and agglomerates contaminants (particularly suspended particles and colloids) in water. Consequently, the aggregates swiftly settle and facilitate easy removal from the water [10]. Coagulants are compounds that are used to remove turbidity and color from raw water [11]. They achieve this by encouraging the production of large agglomerates. In recent years, several studies have been conducted to investigate the safety of a blend of synthetic and natural coagulants that are sustainable and eco-friendly as a replacement for the use of inorganic coagulants alone in the treatment of drinking water [12,13].

Alum is the most commonly used synthetic coagulant in developing countries. It is mainly used because of its excellent turbidity-removal efficiency [14]. Besides its excellent characteristics, it also has a negative impact and can harm the brain and bones [15,16]. To minimize the negative impact, it should be blended with another friendly coagulant, such as watermelon seed. When alum is used as a coagulant, the sludge produced is bulky and non-biodegradable, posing disposal issues and increasing treatment costs [17,18]. The cost of alum is also constantly increasing, making it difficult to afford the fee for this coagulant. However, there has been limited research on mixing a natural coagulant derived from watermelon with a synthetic coagulant (alum) to create a more efficient coagulant that works well with raw surface water. According to Ref. [19], the water purification process for drinking purposes uses natural coagulants derived from plants. Watermelon (*Citrullus lanatus*) can grow in the field by either planting seeds or using containerized transplants. Because of their adsorbent qualities, watermelon seeds can effectively purify water [10,20].

Ethiopians do not use watermelon seeds as food, people eat the main fruit and then discard the seeds. Watermelon is one of the fruits used to make juice. It is locally available, so it is a wise decision to use this precious fruit seed for water treatment. Therefore, it is necessary to reduce the amount of alum in water by incorporating another bio-friendly coagulant into the mixture [21]. To determine the extent of water pollution, one must check the physicochemical parameters of surface water, which contain a variety of pollutants. Most conventional treatment procedures have several variables. To maximize the treatment performance, optimization plays a crucial role [22–24]. Response surface methodology (RSM) is one of the optimization methods that is used to provide a great deal of data [21, 25–27]. It is a set of mathematical and statistical approaches for constructing models, analyzing the impacts of multiple variables, and determining the values of process variables that result in desirable response values [28–30]. The main goal of RSM is to obtain the system's optimal operational conditions or to acquire a place that meets the operating specifications. The objective of this study is to investigate the efficiency of blended natural and synthetic coagulants for surface water treatment. The physicochemical parameters of surface water, specifically COD, BOD, and TDS, were characterized both before and after treatment.

2. Materials and methods

2.1. Materials

The study was conducted in the Oromia region, Jimma zone, Jimma town, near the town water treatment plant facility, and particularly at the intake structure constructed alongside the Gibe River. It is located at 7° 38′ 51″ N latitude and 36° 52′ 12.5″ E longitude. The apparatus used was a COD reactor (Hatch 45600-02), COD kit, heaters (HT-14), pH meter, spectrophotometer (OHSP-350) standard flasks (FSK 20), Erlenmeyer flasks, drying oven (OV-43), measuring cylinder, plastic bottles, burettes, thermometer, spectrophotometer (model 6700), wash bottle weighing balance (model Pw −124), graduated cylinder meter (pH 3310) and conductivity meter (Cond 3110) were used in this study. Chemicals used in surface water treatment and analyses were mercuric sulfate (98.5 %), sodiumhydroxide (98), alum (98 %), silversulfate solution (99.8 %), phenoldisulfonic acid (98.4 %), ammonium hydroxide (98.2 %), EDTA reagent (98.5 %), phenolphthalein indicator, ammonium molybdate (98 %), silver sulfate (97.8 %), stannous chloride (98.8 %), hydrochloric acid (35 %), stock phosphate solution (98.2 %), potassium dichromate solution (97.6 %), sulfuric acid (98.08 %), ferrous ammonium sulfate (FAS)(98.5 %), distilled water and potassium hydroxide (85 %).

2.2. Methods

2.2.1. Study design

The study design comprises material and sample collection, collection of watermelon seed from the juice bar, washed with distilled water then dried with the sun, extract of the oil from powder by using, collection of alum from the laboratory, treatment of the water as per the design parameters, test and record data for all results, characterization and optimization by using RSM, justification about optimum working parameters and conclusion and recommendation.

2.3. Preparation of coagulant

2.3.1. Extraction of natural coagulant

The seeds of watermelon (Fig. 1a) were collected. The seeds are washed thoroughly with distilled water to remove any dirt, debris, or impurities that might affect the extraction process. The sun-dried seeds are crushed into a fine powder using a mortar and pestle. A finer powder increases the surface area, improving solvent contact during extraction. Oil extraction is a step that can enhance the effectiveness of watermelon seed powder as a coagulant by removing a potential interfering substance. Hence, the powdered watermelon seeds were placed in a Soxhlet apparatus (Fig. 1b), and n-hexane, a solvent suitable for oil extraction, was used to selectively dissolve the oil from the seeds. The temperature was maintained at 38 °C. After oil extraction, the remaining seed material (the "cake") was washed with distilled water to remove any residual n-Hexane. This ensures the final product is free of solvent traces. The washed cake is then dried in an oven at 40 °C. This relatively low temperature helps preserve the coagulant properties in the seed powder. Drying continues until a constant weight is achieved, indicating all the moisture has evaporated. Finally, the dried cake was sieved to the size of 150 μm to obtain a fine powder. This fine powder is the natural coagulant extracted from the watermelon seeds.

2.3.2. Alum preparation

In this study, while different types of alum exist, aluminum sulfate ($Al_2(SO_4)_3$) was the preferred type for wastewater treatment. To keep the consistency with watermelon seed powder, alum was sieved to the size of 150 μm to obtain a fine powder.

2.3.3. Blended natural and synthetic alum

In this study, different composition of watermelon seed coagulant was mixed at different ratios of synthetic alum. All ratios investigated were three watermelon seed powder to alum ratios (1:1,1:3 and 3:1).

The laboratory-based (experimental) study was carried out in the Environmental engineering laboratory room at Jimma University, with a focus on the treatment efficiency of blended natural and synthetic coagulants for surface water in the case of Gibe River.

2.4. Sample collection, transportation, and preservation

I. Sample collection

The sample water (Fig. 2) was taken from Jimma town, Boye water treatment plant, at the intake before entering the treatment facility, by plastic Jerrycans. Samples were taken at different three shifts (morning, afternoon, and about the evening), two jerrycans were used at each shift (one for reserve) and a total of 60 L were ready for the run (one jerrycan from each shift), the samples were mixed to have more reliable and representative data.

II. Sample transportation and preservation

The water sample taken from the intake was immediately transported to the laboratory, and the basic parameters temperature and pH were measured immediately to keep the original characteristics [31] of the sample, these parameters were checked in each treatment and tested before carrying out the process.

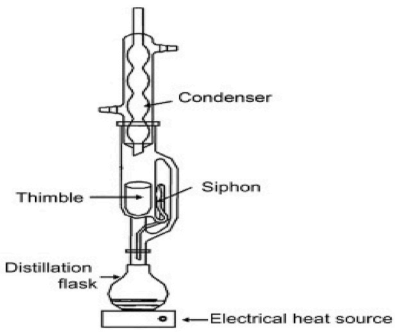
2.5. Experimental setup

After recording the temperature, conductivity, and pH of the sample water, the treatment was carried out by jar test. It consists of six identical 2-liter beakers, each with a stirrer, that can be operated simultaneously. Because the stirrers are powered by a single motor, they all rotate at the same speed [31–33]. The experimental setup is illustrated in Fig. 3.

2.6. Laboratory procedures

All necessary procedures were followed carefully while conducting this study, and the manual for the entire procedure was obtained from the environmental engineering laboratory. Three watermelon seed powder-to-alum ratios (1:1,1:3 and 3:1) were considered. The dosage of blended coagulant i.e. watermelon seed coagulant to alum ranges from 0.1 g/l to 0.7 g/l. The settling time

a)



b)

Fig. 1. Watermelon seed (a) and Soxhlet extractor (b).

Fig. 2. Taking a sample from the intake of Jimma town water treatment plant.

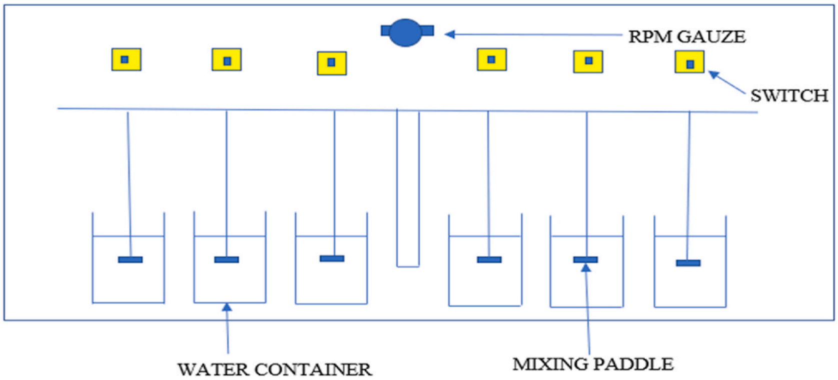


Fig. 3. Jar test setup.

ranges between 20 and 60 min and the contact time is in the range of 5–25 min.

2.7. *Study variables*

The dependent variables of the treatment process were the percentage removal of COD, BOD, and TDS. The independent variables are pH, dosage of coagulants, contact time, and settling time.

2.8. *Method of data analysis and presentation*

After the data had been collected effectively, the gathered data was analyzed and interpreted using Microsoft Excel Office and Response Surface Methodology (RSM) software. It was optimized and analyzed using both qualitative and quantitative data analysis techniques.

2.8.1. *Analysis by empirical formula*

The performance of the process was evaluated based on the responses of COD, BOD, and TDS removal efficiencies as shown in Equation (1), Equation (2), and Equation (3).

I. Percentage of Chemical Oxygen Demand (COD) removal

But,

$$COD\left(\frac{mg}{L}\right)=\frac{Volume\ of\ blank-Volume\ ofsample*N*8}{Volume\ of\ sample}*1000 \tag{1}$$

Where: N- Normality.

II. Percentage of TDS removal

$$Percentage\ removal\ of\ TDS = \frac{TDSi - TDSf}{TDSi}*100 \quad (2)$$

Where:

TDS_i = the initial total dissolved solid in mg/L.

TDSf = the final dissolved solid in mg/L.

III. BOD removal

$$Percentage\ removal\ of\ BOD = \frac{BODi - BODf}{BODi}*100 \quad (3)$$

Where:

BODi: Influent BOD (Biochemical Oxygen Demand). This is the amount of oxygen dissolved in water that microorganisms need to break down organic matter over a specific period, usually 5 days (BOD5). It's measured in milligrams per liter (mg/L).

BODf: Effluent BOD. This is the BOD remaining in the wastewater after treatment. It's also measured in mg/L.

2.8.2. *Experimental design and statistical analysis*

Design-Expert (11.0) software was used to analyze the experimental results. Response surface methodology is the best technique used for the optimization process of the independent variable [34–36]. It is also used to determine the factor which affects the yields of ethanol in a better manner. It helps to determine which factor is significantly affecting the yield and insignificantly affects the yields [3,37,38]. In this response surface methodology, central composite design (CCD) was selected to determine linear (one-factor effect), interaction (multiple-factor effect), and quadratic of the independent variable during the treatment of wastewater using naturally blended coagulants. Response surface methodology generates actual data points, axial data points, and center data points [21,29,39]. This helps to predict the interaction effects of each factor; the data points were selected based on different works of literature and the selected ranges were highly affected by range of pH. In this study, four factors were independent variables and three factors were dependent variables. By using CCD, for 4-factor with three dependent variables, 30 data points were generated based on equation (4) including 6 points as a cent er[40,41].

$$N = 2^n + 2n + Cp \quad (4)$$

Table 1
One-to-one (1:1) removal efficiency of the blended coagulant (WM: Al).

Run	A	B	C	D	COD	BOD	TDS
	–	g/l	min	min	%	%	%
1	9	0.55	30	20	90.74	89.75	94.81
2	3	0.40	40	15	98.74	97.75	98.11
3	7	0.40	60	15	95.68	93.68	96.11
4	9	0.25	50	20	90.14	89.15	95.37
5	7	0.40	40	15	91.22	90.23	94.91
6	5	0.55	50	10	98.08	97.08	97.71
7	9	0.25	50	10	89.22	88.23	93.09
8	11	0.40	40	15	85.62	84.63	91.71
9	5	0.25	30	10	95.74	94.75	95.04
10	5	0.25	50	10	97.32	96.33	96.71
11	7	0.40	20	15	91.14	90.15	92.86
12	7	0.10	40	15	90.22	89.23	92.61
13	7	0.40	40	15	92.24	91.24	94.94
14	5	0.25	30	20	97.67	96.08	96.46
15	7	0.40	40	15	92.72	91.72	94.07
16	7	0.40	40	5	92.65	91.66	92.94
17	7	0.40	40	15	92.22	91.23	94.89
18	5	0.55	30	10	95.68	94.68	96.49
19	7	0.70	40	15	94.14	92.15	96.66
20	9	0.55	50	10	91.34	90.35	94.06
21	9	0.25	30	20	90.88	90.08	94.57
22	7	0.40	40	15	92.22	91.23	95.83
23	9	0.55	50	20	93.14	92.15	93.86
24	9	0.55	30	10	91.66	90.27	92.21
25	9	0.25	30	10	88.09	87.10	90.46
26	5	0.55	50	20	98.14	97.15	97.71
27	7	0.40	40	15	92.69	91.70	96.81
28	5	0.25	50	20	97.68	96.68	95.57
29	5	0.55	30	20	96.14	95.15	97.86
30	7	0.40	40	25	95.24	94.84	96.86

Where, A = pH, B = Dosage, C= Settling time and D = Contact time.

$N = 2^4 + 2*4 + 6 = 30$ (for one ratio)
$N = 30$ runs $* 3 = 90$ (for three ratio)
Where N = Total number of runs (for the three ratios)
n = Number of independent variables.
C_p = Center point.

3. Results and discussion

3.1. Removal efficiency of a blend of alum and watermelon seed powder on Gibe river

Different physical, chemical, and biological characteristics are found in the Gibe River and therefore they are treated using a blend of alum and watermelon seed powder in three ratios. COD, BOD, and TDS are the main characteristics which are shown in Table 1. Natural and human activities alter water quality parameters in different ways. Sediment is a major cause of water pollution because it can naturally enter watercourses, causing color, turbidity, and other problems with drinking water and acting as a vector for other pollutants that bind to soil particles. Nitrate and Phosphate are naturally occurring nutrients and sometimes human activities increase their concentration in rivers. The removal efficiency of alum watermelon seed powder is shown in Fig. 4 in comparison with before and after treatment.

3.1.1. Removal efficiency of alum and watermelon seed powder at different proportions

In this study, three watermelon seed powder to-alum ratios were investigated. Among the three watermelon seed powder to alum ratios (1:1,1:3 and 3:1), the maximum removal efficiency was obtained at a 1:3 ratio, percentage removal of the coagulants at this ratio for COD, BOD, and TDS was 95.26 %, 96.10 % and 98.26 % respectively. All these conditions were obtained at pH = 3, dosage = 0.4 g/l, settling time = 40 min, and contact time = 15 min. Combining natural and chemical coagulants offers significant advantages in water treatment. Natural coagulants excel at destabilizing smaller particles, while chemical coagulants effectively bridge larger ones. Together, they create larger, stronger flocs faster, improving sedimentation and filtration. Moreover, they can target a wider range of contaminants, from turbidity to heavy metals, often requiring lower overall chemical doses, making the process more efficient and environmentally friendly. A 3:1 ratio of natural to chemical coagulants leads to worse performance due to an excessive amount of natural coagulant could lead to over-destabilization of particles, resulting in smaller, less compact flocs that are difficult to remove. Coagulation performance drops with pH because, most coagulants, such as aluminum sulfate and ferric chloride, rely on hydrolysis to form insoluble hydroxide precipitates. These precipitates act as a bridge between particles, facilitating coagulation. However, at extreme pH levels (too high or too low), the hydrolysis process is hindered, reducing precipitate formation. The interaction that exists between natural and chemical coagulants is complex but generally involves Complementary action and Charge neutralization mechanisms. Natural coagulants often excel at destabilizing smaller particles, while chemical coagulants are more effective on larger ones. Together, they can destabilize a wider range of particle sizes. Both types of coagulants (natural and chemical) can neutralize the surface charge of particles, making them more susceptible to aggregation. The experimental results are presented in Table 1, Tables 2, and Table 3 for 1:1,1:3 and 3:1 ratios of watermelon seed to alum respectively.

Table 1 details the effectiveness of a blended coagulant for water treatment. This coagulant combines watermelon seed powder and alum in a one-to-one ratio. The table likely presents removal efficiency percentages for various pollutants. The passage specifically mentions COD (organic matter), BOD (oxygen demand from organic breakdown), and TDS (total dissolved solids). The results are promising, with all three pollutants exhibiting maximum removal efficiencies exceeding 97.5 %. This suggests that the combination of watermelon seed powder and alum proves to be a highly effective treatment method in removing these contaminants from the water.

Table 2 shows the maximum removal efficiency for COD, BOD, and TDS is 95.26 %, 96.10 %, and 98.26 % respectively at the blended ratio of one to three (watermelon seed to alum). The highest removal efficiency was also obtained at this proportion, this is due to the typical characteristics of alum to remove turbidity, color, and TDS [42] and the COD and BOD removal efficiency of watermelon

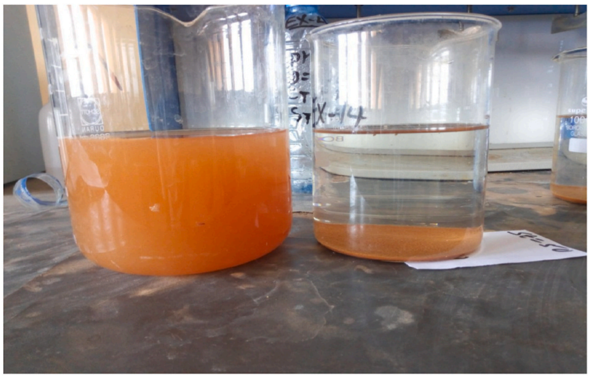


Fig. 4. Sample water before and after treatment.

Table 2
One to three (1:3) ratio removal efficiency of the blend coagulant (WM: Al).

Run	A	B	C	D	COD	BOD	TDS
		g/l	min	min	%	%	%
1	9	0.55	30	20	91.33	91.98	95.81
2	3	0.40	40	15	95.26	96.10	98.26
3	7	0.40	60	15	94.26	96.10	98.26
4	9	0.25	50	20	90.73	91.38	96.37
5	7	0.40	40	15	91.81	92.46	95.91
6	5	0.55	50	10	98.67	99.32	98.71
7	9	0.25	50	10	89.81	90.46	94.09
8	11	0.40	40	15	86.21	86.86	92.71
9	5	0.25	30	10	96.33	96.98	96.04
10	5	0.25	50	10	97.91	98.56	97.71
11	7	0.40	20	15	90.73	91.38	93.46
12	7	0.10	40	15	90.81	91.46	94.01
13	7	0.40	40	15	92.83	93.48	95.94
14	5	0.50	30	20	98.31	93.48	97.46
15	7	0.40	40	15	93.31	93.96	95.07
16	7	0.40	40	5	93.24	93.89	93.94
17	7	0.40	40	15	92.81	93.46	95.89
18	5	0.55	30	10	96.27	96.92	97.49
19	7	0.70	40	15	93.73	94.38	97.66
20	9	0.55	50	10	91.93	92.58	95.06
21	9	0.25	30	20	90.67	92.32	95.57
22	7	0.40	40	15	92.81	93.46	97.83
23	9	0.55	50	20	93.73	94.38	94.86
24	9	0.55	30	10	92.25	92.5	93.21
25	9	0.25	30	10	88.68	89.33	92.46
26	5	0.55	50	20	98.73	99.38	98.71
27	7	0.40	40	15	93.28	93.93	97.81
28	5	0.25	50	20	98.27	98.92	96.57
29	5	0.55	30	20	96.73	97.38	98.86
30	7	0.40	40	25	95.83	97.08	97.86

[43]. the blended coagulant (watermelon seed powder and alum) likely achieves a synergistic effect, improving overall removal efficiency for various pollutants.

Table 3 shows the removal efficiency of blended coagulant in the one-to-one ratio (watermelon seed powder: alum). The maximum removal efficiency for COD, BOD, and TDS is 97.96 %, 96.76 %, and 97.17 % respectively. Both alum and watermelon seed powder can still be removed at different proportions.

3.1.2. *ANOVA for quadratic model for one-to-three ratio (watermelon seed to alum)*

A. COD

From Table 4, the Model F-value of 22.64 implies the model is significant. There is only a 0.01 % chance that an F-value this large could occur due to noise. P-values less than 0.0500 indicate model terms are significant. In this case; A, B, C, D, AB, and D^2 are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. Non-significant lack of fit is good.

B. BOD
C. TDS

From Tables 5 and 6, the Model F-value of 7.98 implies the model is significant. There is only a 0.01 % chance that an F-value this large could occur due to noise. P-values less than 0.0500 indicate model terms are significant. In this case, A, B, C, and D are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. Non-significant lack of fit is good.

3.1.3. *Fit statistics for one-to-three ratio*

From Table 7, the predicted R^2 of 0.7597 is in reasonable agreement with the Adjusted R^2 of 0.9126; i.e., the difference is less than 0.2. Adeq Precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. ratio of 18.807 indicates an adequate signal which is illustrated in Table 7. This model can be used to navigate the design space.

From Table 8, the predicted R^2 of 0.7607 is in reasonable agreement with the Adjusted R^2 of 0.9129; i.e., the difference is less than 0.2. **Adeq Precision** measures the signal-to-noise ratio. A ratio greater than 4 is desirable. A ratio of 18.720 indicates an adequate signal. This model can be used to navigate the design space.

From Table 9, the predicted R^2 of 0.5759 is in reasonable agreement with the Adjusted R^2 of 0.7711; i.e., the difference is less than

Table 3
Three to one (3:1) ratio removal efficiency of the blend coagulant (WM: Al).

Run	A	B	C	D	COD	BOD	TDS
		g/l	min	min	%	%	%
1	9	0.55	30	20	89.96	88.76	93.37
2	3	0.40	40	15	97.96	96.76	97.17
3	7	0.40	60	15	93.89	92.69	93.67
4	9	0.25	50	20	89.36	88.16	94.93
5	7	0.40	40	15	90.43	89.24	93.47
6	5	0.55	50	10	97.29	96.09	96.27
7	9	0.25	50	10	88.43	87.24	91.64
8	11	0.40	40	15	84.83	83.64	90.27
9	5	0.25	30	10	94.96	93.76	93.60
10	5	0.25	50	10	96.53	95.34	94.27
11	7	0.40	20	15	90.36	89.16	91.01
12	7	0.10	40	15	89.43	88.24	91.57
13	7	0.40	40	15	91.45	90.25	93.50
14	5	0.25	30	20	95.58	95.09	95.01
15	7	0.40	40	15	91.93	90.73	92.63
16	7	0.40	40	5	91.86	90.07	91.50
17	7	0.40	40	15	91.44	90.24	93.44
18	5	0.55	30	10	94.89	93.69	95.04
19	7	0.70	40	15	92.36	91.16	95.21
20	9	0.55	50	10	90.56	89.36	92.61
21	9	0.25	30	20	89.29	89.09	93.13
22	7	0.40	40	15	91.44	90.24	94.38
23	9	0.55	50	20	92.36	91.16	93.41
24	9	0.55	30	10	90.08	89.28	90.57
25	9	0.25	30	10	87.30	86.11	89.01
26	5	0.55	50	20	97.36	96.16	96.27
27	7	0.40	40	15	91.90	90.71	94.37
28	5	0.25	50	20	96.89	95.69	94.13
29	5	0.55	30	20	95.36	94.16	96.41
30	7	0.40	40	25	94.45	93.85	94.41

Table 4
ANOVA for quadratic model for COD.

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	302.63	14	21.62	22.64	<0.0001	significant
A-pH	253.31	1	253.31	265.28	<0.0001	
B-Dosage	9.54	1	9.54	9.99	0.0065	
C-Settling time	14.46	1	14.46	15.14	0.0014	
D-Contact time	5.49	1	5.49	5.75	0.0300	
AB	5.56	1	5.56	5.82	0.0291	
AC	0.5650	1	0.5650	0.5917	0.4537	
AD	0.1010	1	0.1010	0.1058	0.7495	
BC	0.7225	1	0.7225	0.7566	0.3981	
BD	0.7560	1	0.7560	0.7917	0.3876	
CD	0.0000	1	0.0000	0.0000	0.9969	
A^2	1.06	1	1.06	1.10	0.3098	
B^2	0.1388	1	0.1388	0.1453	0.7084	
C^2	1.76	1	1.76	1.84	0.1950	
D^2	11.11	1	11.11	11.64	0.0039	
Residual	14.32	15	0.9549			
Lack of Fit	12.86	10	1.29	4.38	0.0583	not significant
Pure Error	1.47	5	0.2933			
Cor Total	316.95	29				

0.2. Adeq Precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. the ratio of 11.833 indicates an adequate signal. This model can be used to navigate the design space.

3.2. One factor effect for one to three ratio

3.2.1. Effect of pH

pH is critical in the coagulation-flocculation process since coagulation occurs within a specific pH range for the coagulant [44]. From the findings, the best optimum removal efficiency was obtained at a dosage of 0.4 g/L, a settling time of 40 min, and a contact

Table 5
ANOVA for quadratic model for BOD.

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	299.44	14	21.39	22.71	<0.0001	significant
A-pH	247.50	1	247.50	262.82	<0.0001	
B-Dosage	8.21	1	8.21	8.71	0.0099	
C-Settling time	13.99	1	13.99	14.86	0.0016	
D-Contact time	7.91	1	7.91	8.40	0.0110	
AB	3.74	1	3.74	3.97	0.0649	
AC	0.9539	1	0.9539	1.01	0.3301	
AD	0.5518	1	0.5518	0.5859	0.4559	
BC	1.27	1	1.27	1.34	0.2645	
BD	0.8921	1	0.8921	0.9473	0.3458	
CD	0.0778	1	0.0778	0.0826	0.7777	
A^2	0.9566	1	0.9566	1.02	0.3295	
B^2	0.1046	1	0.1046	0.1111	0.7435	
C^2	1.63	1	1.63	1.73	0.2082	
D^2	13.52	1	13.52	14.36	0.0018	
Residual	14.13	15	0.9417			
Lack of Fit	12.66	10	1.27	4.32	0.0601	not significant
Pure Error	1.47	5	0.2933			
Cor Total	313.56	29				

Table 6
ANOVA for quadratic model for TDS.

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	96.08	14	6.86	7.98	0.0001	significant
A-pH	59.94	1	59.94	69.68	<0.0001	
B-Dosage	7.85	1	7.85	9.13	0.0086	
C-Settling time	4.60	1	4.60	5.35	0.0354	
D-Contact time	12.43	1	12.43	14.45	0.0017	
AB	1.91	1	1.91	2.23	0.1565	
AC	0.1312	1	0.1312	0.1525	0.7016	
AD	2.37	1	2.37	2.75	0.1177	
BC	0.0956	1	0.0956	0.1111	0.7435	
BD	0.2256	1	0.2256	0.2622	0.6161	
CD	3.57	1	3.57	4.15	0.0597	
A^2	0.0163	1	0.0163	0.0189	0.8925	
B^2	0.0916	1	0.0916	0.1065	0.7487	
C^2	2.81	1	2.81	3.26	0.0910	
D^2	0.0468	1	0.0468	0.0544	0.8187	
Residual	12.90	15	0.8603			
Lack of Fit	6.40	10	0.6398	0.4917	0.8412	not significant
Pure Error	6.51	5	1.30			
Cor Total	108.99	29				

Table 7
Model summary for % COD removal.

Std. Dev.	0.9772	R^2	0.9548
Mean	93.73	Adjusted R^2	0.9126
C.V. %	1.04	Predicted R^2	0.7597
		Adeq Precision	18.8072

Table 8
Model summary for % BOD removal.

Std. Dev.	0.9704	R^2	0.9550
Mean	94.41	Adjusted R^2	0.9129
C.V. %	1.03	Predicted R^2	0.7607
		Adeq Precision	18.7198

Table 9
Model summary for % TDS removal.

Std. Dev.	**0.9275**	**R^2**	**0.8816**
Mean	96.09	Adjusted R^2	0.7711
C.V. %	0.9652	Predicted R^2	0.5759
		Adeq Precision	11.8327

time of 15 min at a point of 1:3 watermelon seed coagulant to alum ratio. At this optimum point of the removal efficiency, 0.4 g/L of dosage consists of a ratio of 1:3 (watermelon seed coagulant to alum coagulant) i.e watermelon seed coagulant = 0.1 g/L and Alum 0.3 g/L. To investigate the effect of pH increasing its value from 3.0 to 11.0 by keeping other factors constant and this shows a significant effect on the removal efficiency. The maximum removal efficiency was obtained at a pH of 3.0. The removal efficiency for COD, BOD, and TDS was 95.26 %, 96.18 %, and 98.26 % respectively. All response almost decreases sharply as pH increases from 3.0 to 7.0 but increasing pH from 7.0 to 11 slightly affects the removal efficiency which is shown in Fig. 5.

3.2.2. *Effect of dosage*

The removal efficiency of the coagulant varies with the dosage. The amount of coagulant added to water affects the quality of the treated wastewater [45]. Fig. 6 shows that the physicochemical properties of the wastewater changed as the coagulant dose in the water was increased. Increasing the dose of the blended coagulant (watermelon seed to alum 1:3) from 0.1 to 0.7 g/L improves the coagulant's removal efficiency across all responses. As the coagulant was increased to 0.7 g/L the maximum removal efficiency was obtained and the removal efficiency for COD, BOD, and TDS values rose to 93.73 %,94.38 %, and 97.65 % respectively. The findings support the blended coagulant as an effective coagulant. Individually, alum removes suspended solids and other physicochemical properties from river water [14]. The watermelon seed's high protein, tannin, and mineral content also could be responsible for its coagulant properties [10].

3.2.3. *Effect of contact time*

The quality of water is influenced by the duration of contact between the coagulant and the water [10]. During the coagulation process, rapid mixing is employed to evenly distribute the coagulant throughout the water. Conversely, slow mixing is crucial in the flocculation process to achieve optimal outcomes. Sufficient time must be allowed for the production of adequately sized particles that can be efficiently removed during the sedimentation process [46]. The time taken for macro floc formation, also known as flocculation time, is a critical operational parameter in water treatment plants that perform coagulation-flocculation processes.

In the case of a 1:3 ratio (optimum) of watermelon seed to alum coagulant, the highest removal efficiency was achieved with a contact time of 20 min, pH value of 5, dosage of 0.55 g/l, and settling time of 50 min (See Fig. 7). Specifically, the maximum removal percentages were 98.73 % for COD, 99.38 % for BOD, and 98.71 % for TDS at a contact time of 20 min. As the contact time increases, the treatment efficiency also improves. This relationship is depicted in Fig. 10. However, beyond a contact time of 20 min, the removal efficiency reaches a plateau and shows only a marginal decline. This leveling off is attributed to the combined effects of other factors influencing the process.

3.2.4. *Effect of settling time*

The settling time of agglomerates in the treatment basin plays a significant role in determining the overall water quality [42]. The removal percentage for all measured parameters increases as the settling time lengthens, as shown in Fig. 11. However, the rate of removal may differ for each specific parameter. The settling time has a notable and quantifiable impact on the efficiency of the coagulation process. Specifically, the highest removal efficiency was achieved with a settling time of 50 min, a pH value of 5, a dosage

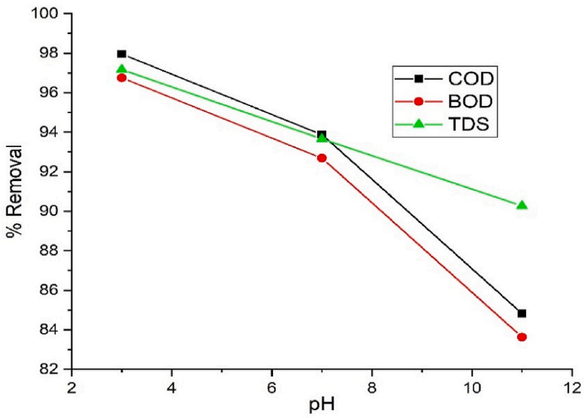


Fig. 5. Effect of pH on coagulation process.

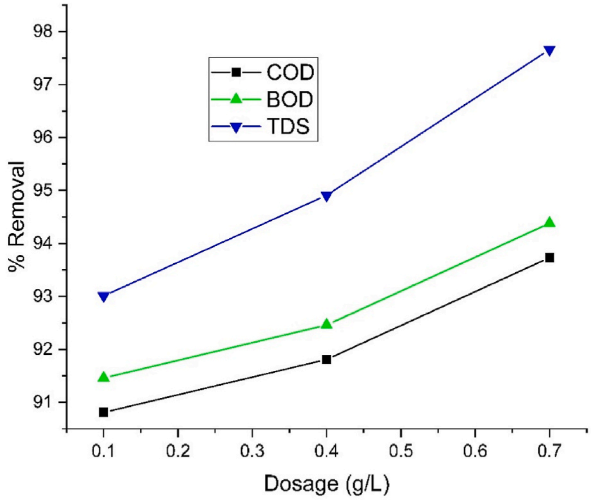


Fig. 6. Effect of dosage on coagulation process.

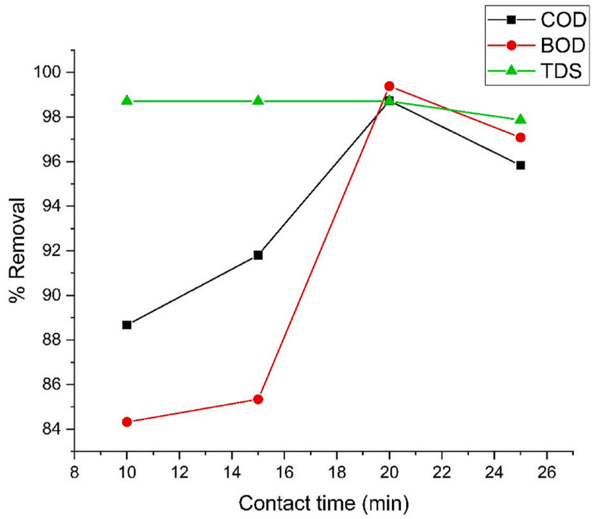


Fig. 7. Effect of contact time on the coagulation process.

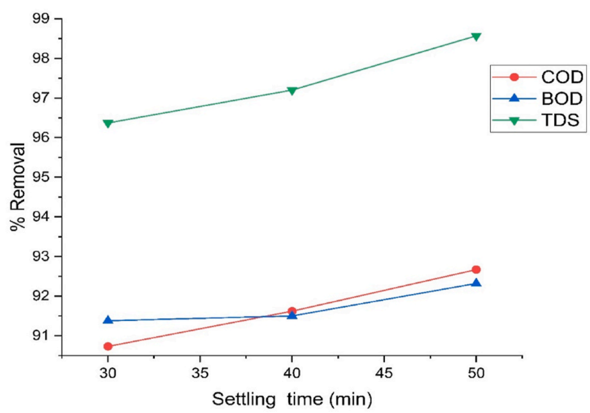


Fig. 8. Effect of contact time on the coagulation process.

of 0.55 g/L (using a 1:3 ratio of watermelon seed to alum), and a contact time of 10 min. The corresponding removal percentages were 98.67 % for COD, 99.32 % for BOD, and 98.71 % for TDS (see Fig. 8).

3.3. *Interactive effects of factors on the responses*

3.3.1. *Effects of pH and dosage*

Fig. 9 presents a 3D graph that reveals the interplay between COD removal efficiency, pH, and dosage. COD removal efficiency, the dependent variable, reflects how effectively the treatment process eliminates organic matter from the water. This organic matter, measured by COD, can negatively impact aquatic life and complicate water purification for drinking purposes. The two independent variables influencing COD removal efficiency are pH (acidity) and dosage (treatment intensity). A color scale on the graph depicts the efficiency; blue: low COD removal efficiency means the treatment isn't effectively removing organic matter. Light green: intermediate COD removal efficiency which means the treatment removes some organic matter, but not optimally. Red: maximum COD removal efficiency which means the treatment process is most effective at removing organic matter. Key insights from Fig. 9 can be summarized as follows; Impact of pH: the graph shows a clear trend – COD removal efficiency is low at a pH of 3 (likely acidic) and increases steadily as the pH gets higher, reaching maximum efficiency around a pH of 11 (likely basic). This suggests that the treatment process relies on chemical reactions that function more effectively under alkaline conditions. At lower (acidic) pH levels, the treatment might be less efficient at breaking down or clumping (flocculating) the organic matter for removal. Impact of Dosage: The graph also reveals a positive correlation between COD removal efficiency and dosage. As the dosage of the treatment chemical (or process intensity) increases from 0.1 to 0.7, the COD removal efficiency increases as well. This indicates that a higher dosage provides more of the necessary chemicals or increases the treatment intensity, leading to more effective removal of organic matter from the water. In essence, Fig. 9 highlights that optimizing COD removal from water requires considering both the water's acidity (pH) and the amount of treatment applied (dosage). The graph suggests that a higher dosage and a slightly basic pH (around 11) are favorable conditions for maximizing COD removal efficiency.

3.4. *Regression equations*

The researchers employed regression analysis to fit a second-order polynomial model (quadratic equation) to the experimental data. This technique helps identify the most relevant factors affecting the response variable — the percentage removal of BOD, COD, and TDS. The independent variables considered in the model are the experimental factors: pH (A), dosage (B), settling time (C), and contact time (D). The final regression equations, likely presented in Equations (5)–(7), express the relationships between these factors in coded terms. Coding simplifies calculations and the interpretation of the coefficients within the equations. These coefficients will indicate the magnitude and direction of the influence each factor (pH, dosage, etc.) has on the percentage removal of pollutants. The analysis establishes an empirical relationship between the response (percentage removal) and the factors affecting it. "Empirical" means this relationship is derived from the observed experimental data rather than theoretical principles. Equations (5)–(7) represent this mathematical description of how the chosen factors influence the effectiveness of the water treatment process. It's important to note that this analysis focuses on identifying the relevant factors and their overall effect on pollutant removal. Further analysis of the equations (e.g., looking at the signs and magnitudes of the coefficients) would be necessary to understand the specific way each factor (pH, dosage, etc.) interacts with the others to achieve optimal removal.

$$COD = 92.81 - 3.25A + 0.6306B + 0.7761C + 0.4782D + 0.5896AB - 0.1879AC + 0.0794AD + 0.2125BC - 0.2174BD \quad (5)$$

$$BOD = 93.46 - 3.21A + 0.5847B + 0.7636C + 0.574D + 0.4833AB - 0.2442AC + 0.1857AD + 0.2812BC - 0.2361DB \quad (6)$$

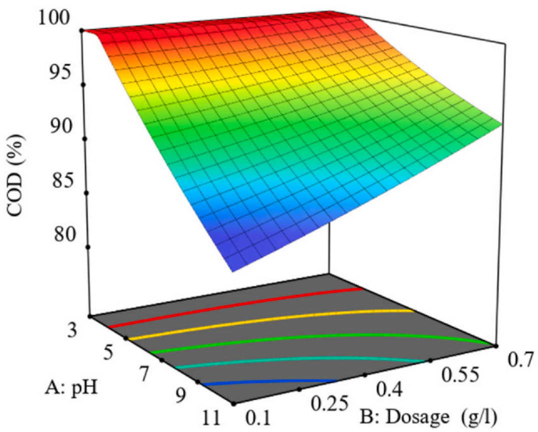


Fig. 9. Interactive effects of pH and dosage on COD.

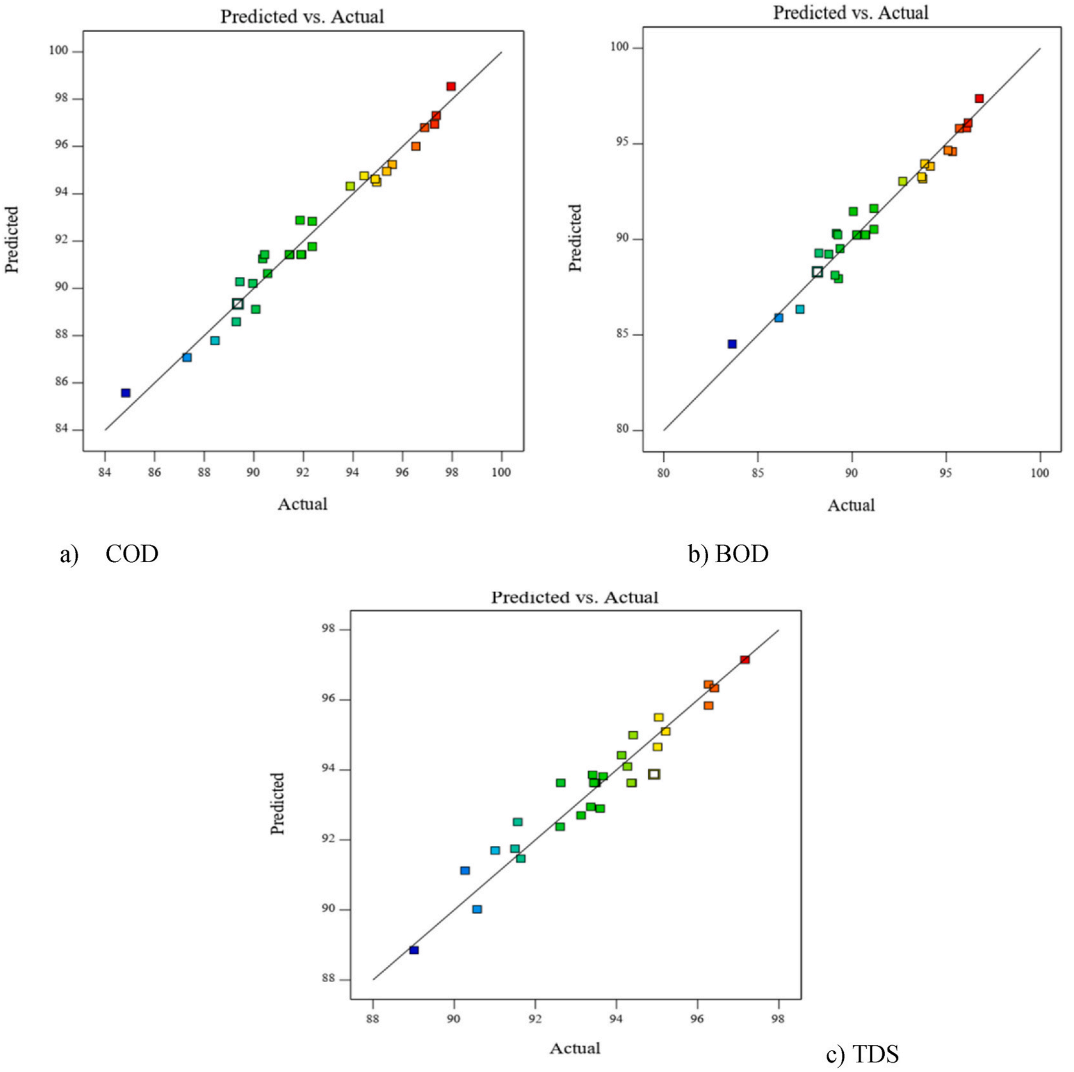


Fig. 10. Actual vs predicted values of the significant models for the optimum conditions a) COD b) BOD c) TDS.

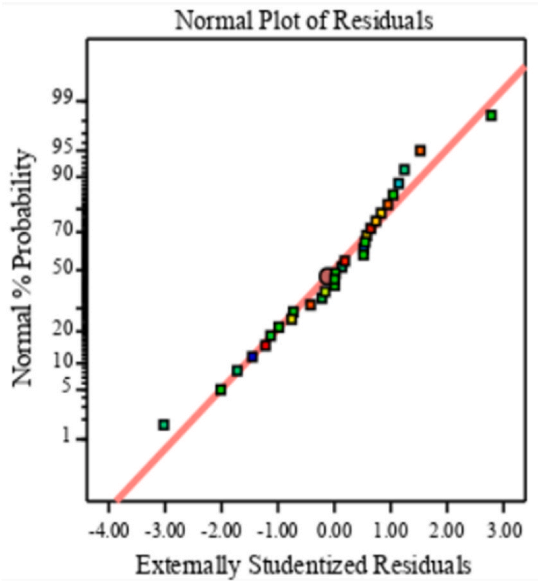
$$TDS = 96.41 - 1.58A + 0.572B + 0.4378C + 0.7196D - 0.3459AB + 0.0906AC + 0.3848AD - 0.0773BC - 0.1187BD \tag{7}$$

According to the findings, all individual factors namely pH (A), Dosage (B), settling time (C), and contact time (D) were significant in all proportions, and many of the interactive factors (AB, AC, BC, BD, CD, A^2, B^2, C^2, and D^2) were significant. The lack of fit test was used to validate the model. On the regression model, for $p < 0.05$ the model is significant and non-significant if $P > 0.05$.

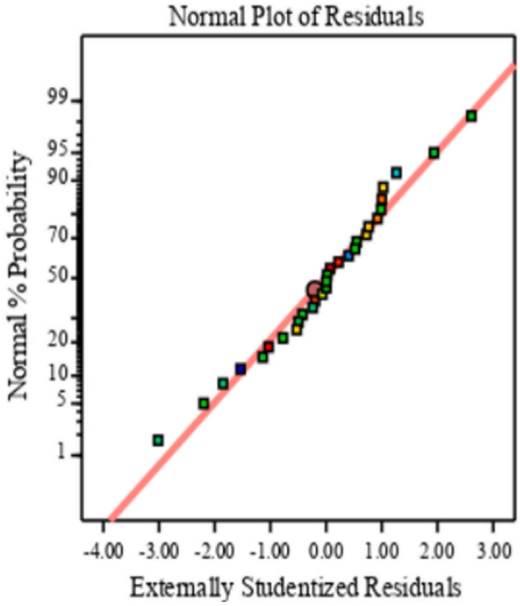
3.5. The maximum condition for responses

Fig. 10 depicts a scatter plot of actual versus predicted values for the responses. Ideally, this plot should show a cluster of points concentrated around a diagonal line, indicating a strong positive correlation between the actual measurements and the model's predictions. Such a pattern suggests the model is reliable in capturing the relationship between the independent variables and the responses. While Fig. 10 focuses on overall model performance, further analysis likely involved examining residuals (the difference between actual and predicted values). This additional analysis, perhaps not shown here, would help identify potential outliers or areas for improvement in the model. The text also mentions the maximum observed response (likely the best water quality improvement) achieved under specific experimental conditions. These optimal conditions, including the watermelon seed powder to alum ratio, pH dosage, settling time, and contact time, are likely detailed in separate tables (Tables 10–15) but not explicitly mentioned in this excerpt.

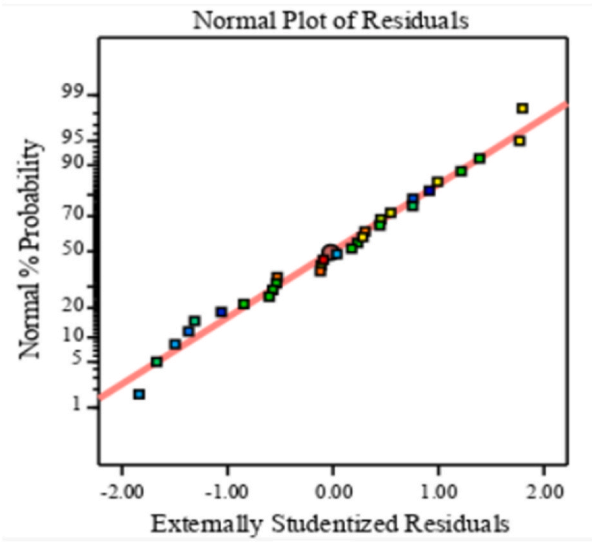
The optimum values of the 3 tables listed above were obtained from the software, and they were selected because of their degree of desirability.



a) BOD



b) COD



c) TDS

Fig. 11. Normal probability plot for (a) BOD, (b) COD, and (c) TDS.

3.6. *Graphical analysis of valid models*

Statistical analysis was conducted to assess water quality parameters like COD, BOD, and TDS. To ensure reliable results, the researchers checked if the errors (residuals) from the analysis followed a normal distribution. This normality check was done using a normal probability plot (NPP), which is visualized in Fig. 11 of the document. An ideal outcome in an NPP is a straight line, and

Table 10
Optimization of coagulation process variables under optimum conditions (1:1).

Ratio		1:1							
Solution number	pH	Dose	St	Ct	% COD	% BOD	% TDS	desirability	Status
1	6.15	0.42	42.6	18.1	94.4	93.4	96.4	1	Selected

Table 11
Optimization of coagulation process variables under optimum conditions (1:3).

Ratio					1:3				
Solution number	pH	Dose	St	Ct	% COD	% BOD	% TDS	desirability	Status
1	5	0.55	50	20	94.62	94.33	98.5	0.918	selected

Table 12
Optimization of coagulation process variables under optimum conditions (3:1).

Ratio				3:1					
Solution number	pH	Dose	St	Ct	% COD	% BOD	% TDS	desirability	Status
1	5	0.55	50	10	90.4	91.4	96.4	0.91	Selected

Table 13
Model validation by experimental results under optimum conditions.

Factors				Percentage removal efficiency		
pH	Dose	S.time	C.time	¯COD	BOD	TDS
6.15	0.42	42.6	18.1			
Model predicted				94.4	93.4	96.4
Experimental				93.1	91.7	90.4

Table 14
Model validation by experimental results under optimum conditions.

Factors				Percentage removal efficiency		
pH	Dose	S.time	C.time	COD	BOD	TDS
5	0.55	5	20			
Model predicted				94.62	94.33	98.5
Experimental				91.1	91.4	94.8

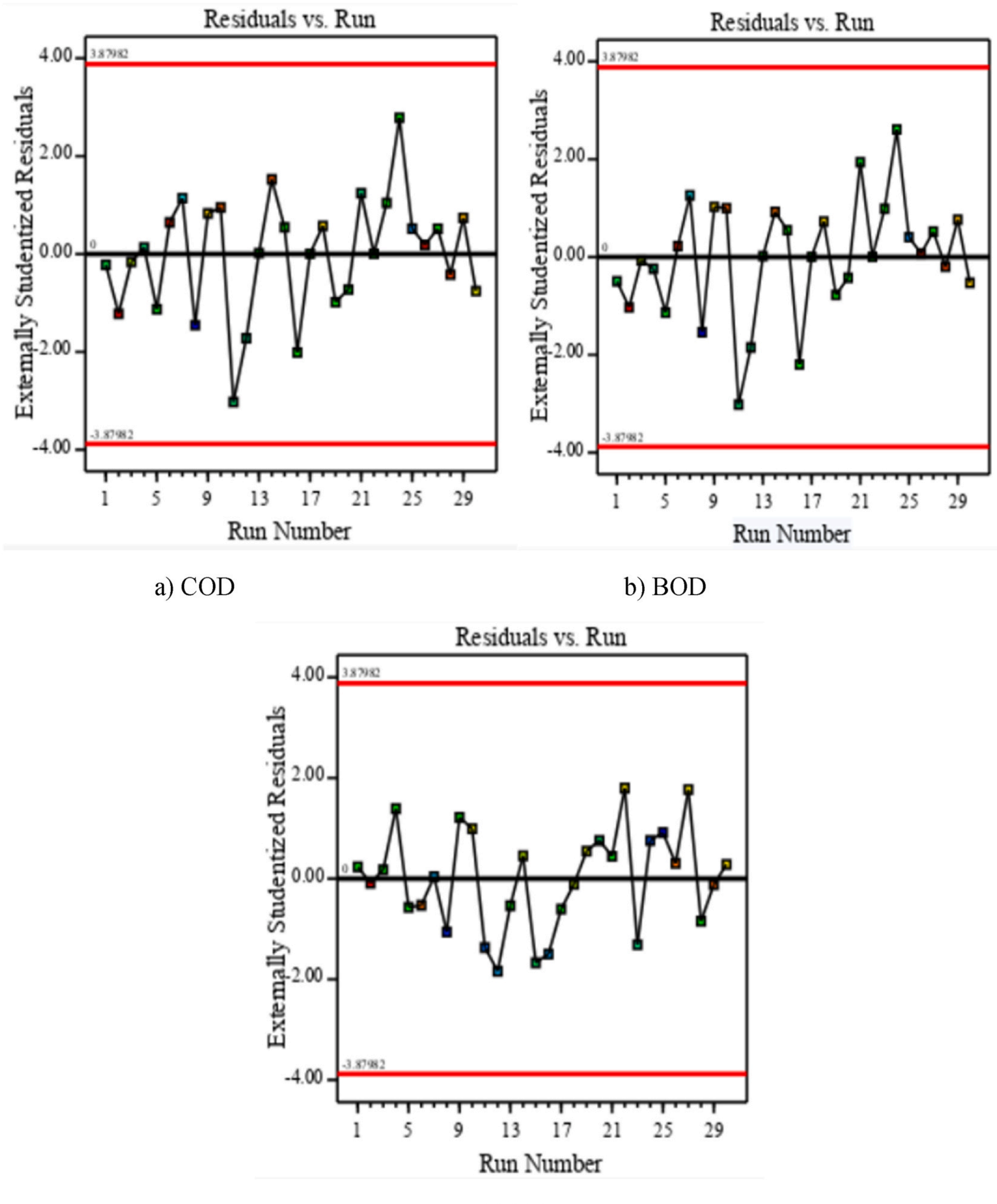
Table 15
Model validation by experimental results under optimum conditions.

Factors				Percentage removal efficiency		
pH	Dose	S.time	C.time	COD	BOD	TDS
5	0.55	50	20			
Model predicted				90.4	91.4	96.4
Experimental				88.7	88.4	92.1

*S.timet: settling time C.time: contact time.

according to the passage, Fig. 11 shows this pattern for all the parameters. This indicates that the residuals are normally distributed, likely due to a mathematical transformation applied to the data beforehand. This transformation, such as converting values to logarithms, improves the normality of the errors, making the overall analysis more trustworthy.

Fig. 12 presents a residual plot for BOD, COD, and TDS, where the residuals are plotted against the order in which the experiments were conducted. Ideally, this plot should exhibit a random scatter of points around the horizontal line (zero residual). This pattern indicates that there are no systematic trends or biases related to the order the experiments were run. The text mentions “two threshold lines” which are likely horizontal lines drawn above and below zero on the residual axis. These lines help identify potentially problematic data points (outliers). Outliers, represented by points falling outside the threshold lines, could indicate errors during a specific experiment or the influence of a factor not considered by the model. In this case, the fact that all points fall within the threshold lines for BOD, COD, and TDS is a positive finding. This suggests that the data likely has no major outliers and the model performs reasonably well across all experiment orders, regardless of the order they were conducted in. It’s important to remember that the absence of outliers in this specific plot is just one aspect of model evaluation. Further analyses are likely needed for a more comprehensive assessment of the model’s overall performance.



a) COD

b) BOD

c) TDS

Fig. 12. Run number vs Residual plot for a) COD b) BOD and c) TDS.

4. Conclusion

Surface water treatment by using a blend of synthetic and natural coagulants is a novel and effective approach that can treat basic parameters of physical, chemical, and biological characteristics. Treatment was done by considering variables as independent and dependent. pH, dosage, settling time, and contact time were independent variables whereas removal efficiency for seven responses (COD, BOD, and TDS) was the independent variable. In this study effects of the independent variables significantly affect the treatment efficiency of the coagulants. To obtain the maximum treatment efficiency, Response Surface Methodology (RSM) software was used. Optimum conditions were selected for three of the three different ratios, beside obtaining the optimum conditions, validation of data and model was also checked through experiment. The central composite design (CCD) was used over the other design due to its relatability behavior. Numerical and statistical data were also analyzed. The confidence interval for the model significance test was taken at 95 %, for fit statistics the differences between Predicted R^2 Adjusted R^2 were kept less than 0.2. Among the three watermelon seed powder to alum ratios (1:1,1:3 and 3:1), the maximum removal efficiency was obtained at a 1:3 ratio, percentage removal of the coagulants at this ratio for COD, BOD, and TDS was 95.26 %, 96.10 % and 98.26 % respectively. All these conditions were obtained at pH = 3, dosage = 0.4 g/l, settling time = 40 min, and contact time = 15 min. This implies that the combination of alum with watermelon seed powder can treat surface water highly for agricultural and domestic purposes by minimizing the negative effect of using synthetic coagulants in terms of health, environment, and economy. A blend of watermelon seed powder and alum as a coagulant is a very important novel approach in municipal water treatment systems due to its compatibility with the socio-economic level of the society and its tendency to minimize the side effects of synthetic coagulants alone on health and environment in developing countries like Ethiopia. Despite its effectiveness as a coagulant, the following factors must be considered. These are: i) Coagulant is effective in acidic water (pH = 3), so it is recommended to apply this test on other plants that generate acidic water as effluent ii) After the treatment of the river water by using the blend coagulant the water remains acidic: therefore, it should be neutralized by using proper basic solution iii) Because most Sub-Saharan countries are comfortable with watermelon, the agro economics sector should develop planting this plant for greater effectiveness iv) Ways shall be searched to separate the biodegradable sludge from nonbiodegradable to recycle the biodegradable sludge to the environment as means of compost. V) Detail cost analysis was not done in this study: therefore, deep cost analysis should be done to check the feasibility of this work.

Data availability

All data are available within the manuscript.

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CRediT authorship contribution statement

Yared Endale: Funding acquisition, Formal analysis, Data curation. **Zerihun Asmelash Samuel:** Methodology, Investigation, Conceptualization. **Seifu Kebede:** Visualization, Software. **Abreham Bekele Bayu:** Writing – review & editing, Writing – original draft, Resources.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Yared Endale reports financial support was provided by Jimma University Institute of Technology.Zerihun Asmelash Samuel and Seifu Kebede reports a relationship with Jimma University Institute of Technology that includes: implementation. Abreham Bekele Bayu has coordination for achievement of the task. All of authors are academic staffs in Jimma Institute of Technology, Ethiopia If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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