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Natural Coagulant Efficiency of Moringa Oleifera Seeds in Raw Water Treatment

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Abstract. Adverse health and environmental impacts of conventional chemical compounds in treating water and wastewater encouraged the study of natural alternatives, especially plant-based compounds. This study aims to investigate the feasibility of Moringa oleifera (MO) seeds as an eco-friendly natural coagulant and to compare their effectiveness with the chemical coagulant – aluminum sulfate (Alum). The performance of MO in turbidity and chemical oxygen demand (COD) removal from raw water employing the jar test was examined. Coagulation conditions were optimized using a jar test to determine the optimum dose of MO and pH level. The impact of the MO on treated water characteristics (electrical conductivity, pH, and total dissolved solids) was also examined. The experiment used a range of MO doses (50, 100, 150, 200, and 300 mg/l) and Alum (5, 10, 20, 30, and 40 mg/l) to treat raw water samples. The performance of both coagulants followed similar trends. The maximum turbidity removal efficiency was about 97 % obtained at an optimum dosage of 100 mg/l of MO, while for COD, the highest removal was about 49 % at 150 mg/l of MO. No significant effect of MO dosing on the pH level of the solution was noticed, while a substantial increase in electrical conductivity from 591 ± 1.5 μS/cm to 620 ± 1.5 μS/cm was observed when the dosage of MO increased to 300 mg/l. Total dissolved solids (TDS) also increased for raw water from 255 ± 1.0 μS/cm to 367 ± 2.0 μS/cm at a 300 mg/l dosage. One-way analysis of variance for the relation between dose and turbidity removal shows that the model is significant (*F*-value of 76.1, *P*-value less than 0.0001).

Keywords: water treatment, purification, coagulation, jar test, turbidity, chemical oxygen demand (COD).

1 Introduction

The increased demand for efficient water treatment techniques encouraged researchers to investigate sustainable and eco-friendly alternatives to conventional chemical coagulants [1]. Recent human activities and development have led to decreased potable water because of increased discharge of pollutants to nature due to rapid economic growth, resulting in annual deaths of more than 2,000,000 children under five from water-borne pathogens [2].

Discharges from different sources decrease the reliability of surface water by flowing and discharging into water streams due to the harmful chemical substances that exist in them, leading to a considerable cause of water pollution [3, 4]. Drinking water quality is specified through different parameters and characteristics, and its continuous examination is quite significant as long-term utilization of polluted water is believed to increase the risk of many diseases [5, 6].

Different physicochemical methods are used for water treatment. Conventional techniques include rapid mixing, coagulation, flocculation, biological processes, adsorption, filtration, ion exchange, and sedimentation. These techniques may be applied alone or by combining multiple approaches, depending on the degree of water contamination and the required water quality [7, 8]. Adsorption can effectively eliminate dissolved matter, but it can be expensive for large water volumes. Replacement or regeneration of adsorbents is essential, adding to the cost of the treatment method. Filtration is prone to clogging, making preceding coagulation crucial to treatment processes. Elevated organic and suspended materials concentrations in water will raise water turbidity, acting as transmitting media for disease-causing microorganisms, so, removing the turbidity is a significant stage in water treatment, Water turbidity removal accomplished by techniques like coagulation due to its simplicity, cost-effectiveness, low energy demand, high efficiency and better sludge handling if compared to other processes, Moreover, coagulation is often used as a preliminary step to improve the performance of secondary and tertiary treatments [9, 10], which mainly achieved by addition of chemicals, Alum for example [11].

Coagulation and flocculation are an essential part of the water treatment process. Coagulation is achieved by adding compounds (artificial or natural) to the water to neutralize the negative charges of suspended materials in water [12]. Flocculation is a slow, mild water stirring to enhance the flocs to build up to a higher size for effective settling [13]. The removal efficiency is related to the coagulant dose. Aluminum sulfate (Alum) is used extensively in coagulation due to its potential and low cost. Conversely, it generates by-products that affect an individual's health [14, 15].

The chemical coagulation process may generate a considerable amount of toxic sludge. Alum has been implicated as a possible cause of neurological illnesses [16, 17]. It is widely used in the coagulation process through the water treatment process; meanwhile, it may lead to chronic health effects [18, 19]. When aluminum enters the human body, bone and brain damage may occur, and it is characterized as a neurotoxic substance. The generated sludge contains a high percentage of non-biodegradable aluminum compounds, increasing the treatment cost [20].

MO seeds have become a promising natural coagulant for treating water and wastewater, demonstrating high turbidity and microbial removal. Studies show that the seeds include water-soluble, positively charged proteins in their composition, which effectively act as coagulants by destabilizing suspended particles, offering a biodegradable and cost-effective alternative to chemical coagulants [21, 22].

2 Literature Review

Herrera et al. [23] used two coagulants obtained from Moringa oleifera (MO) seeds (powder and extract) against a traditional coagulant, Alum, with various dosages to evaluate their effectiveness in reducing wastewater turbidity. The study found that both coagulants derived from MO seeds were highly effective in reducing turbidity in wastewater, achieving removal of 97 % and 99 %, respectively. The powder form required a lower dosage, with higher sedimentation periods than the extract.

Nzeyimana and Mary [12] studied MO as a natural coagulant resulted in high removal in many water quality parameters, such as turbidity (92 %), chemical oxygen demand (COD) (88 %), total solids (96 %), chloride (75 %), total hardness (74 %), and inorganic phosphorous (68 %), the optimum dose of 400 mg/l was applied. However, the specific reduction in biochemical oxygen demand (BOD) was not calculated in the study, showing a gap in the assessment of the efficacy of MO in the treatment of this critical water quality parameter, which necessitates further investigation.

A study conducted by Ariani et al. [19] found that using MO seed powder as a coagulant significantly removed the contaminants in industrial effluent; the optimal conditions were a mixing time of 3 minutes and a coagulant dosage of 4 g/l. The COD reduction efficiency was 65 %, while for total suspended solids (TSS) and turbidity, the reduction efficiency of 53 % for TSS and 59 % for turbidity, showing the potential of MO seeds in treating industrial discharge, on the other hand, The research does not investigate the comparative effectiveness of Moringa seed powder against other conventional coagulants or natural coagulants, which could provide insights into its relative performance and practicality in various wastewater treatment scenarios.

In the research conducted by de Oliveira et al. [24], the coagulant was prepared in saline solutions (1 M NaCl and 1 M KCl) immediately before the experiments to ensure freshness and effectiveness during the coagulation/flocculation process. The study found that the ideal coagulant doses, Moringa-NaCl and Moringa-KCl, were 1.0 g/l and 1.5 g/l. The reliance on specific saline solutions (NaCl and KCl) for preparing the coagulant, which may not be readily available or practical in all settings, may limit the applicability of the findings in diverse geographical or economic contexts.

Sinaga et al. [25] studied Moringa seeds powder as a natural coagulant for treating Batik effluents, with varying doses of 2, 6, 10, 14, 18, and 22 g/l to analyze its effectiveness in reducing TSS and BOD concentrations, the concentration of total suspended solids (TSS) in Batik wastewater decreased significantly by approximately 84 % when using Moringa seeds with optimum dose of 18 g/l.

The study by Padilla et al. [26] involved collecting MO grains, which were then subjected to cold oil extraction using a mechanical press. The resulting cake was prepared for a natural coagulant through saline extraction with sodium chloride. The study found that natural coagulant derived from MO cake demonstrated similar color and turbidity reduction when compared to the chemical coagulant Alum; the chemical coagulant caused a significant decrease in pH, and the natural coagulant did not show a statistically significant difference in pH.

A new method of preparing a coagulant from MO seeds, suggested by Hounsinou et al. [27], includes: collecting the seeds, peeling, grinding the seeds, oil and salts extraction from the powder, filtration with a 0.2 μm filter to obtain the coagulant in dry form. The new coagulant produced from MO seeds effectively reduced the turbidity of lake water from an initial 40 NTU (nephelometric turbidity unit) to almost 5 NTU, achieving a reduction of nearly 95 % when using just 0.7 mg/l. However, the study does not explore the synthesized coagulant's long-term stability and storage conditions, which are critical for practical applications in water treatment.

The research achieved by Rifi et al. [28] which investigates the treating of olive oil mill wastewater by flotation and coagulation procedure, specifically employing MO seeds as a natural coagulant to reduce contamination levels in the effluents, resulted in significant reductions in various pollution parameters: polyphenols were decreased by 66 %, turbidity by 83 %, COD by 58 %, suspended solids by 75 %, and nitrates by 73 %. The study does not explore the chronic effects of using this coagulant on the environment or the potential accumulation of by-products in the ecosystem after treatment, and lacks a comprehensive comparison with other conventional treatment methods.

The use of MO seed as a natural coagulant for treating oily steelworks wastewater was discussed by Lester-Card et al. [29], highlighting its effectiveness in reducing turbidity and oil content in the effluent. The study demonstrated that MO seed extract reduces turbidity and oil content in oily steelworks wastewater, indicating its potential in wastewater treatment and significant improvement in water quality characteristics.

This study aims to investigate the feasibility of MO seeds prepared employing simple steps to be used as an eco-friendly natural coagulant and to compare their effectiveness with a chemical coagulant (Alum). We were also interested in optimizing coagulation conditions to determine the optimum dose of MO. A series of jar tests was conducted using MO to evaluate its effectiveness in removing turbidity and COD from raw water samples. Jar test experiments were also performed using Alum to compare its efficiency with MO in turbidity removal. The influence of MO on the treated water characteristics, including electrical conductivity, total dissolved solids (TDS), and pH, was also investigated. This study demonstrates MO as a part of enhancing sustainability while addressing the challenges of efficient water treatment technologies.

3 Research Methodology

3.1 Coagulant preparation and stock solution

MO were peeled, rinsed with distilled water, and then dried at room temperature, as shown in Figure 1.

a b

Figure 1 – Moringa seeds before (a) and after (b) grinding

Grinding the seeds was achieved by an industrial grinder. The powdered sample was sieved through sieve no. 40 (with 425 μm openings), then stored in a dry container.

1 g of the powder was added to 200 ml of distilled water to achieve a 5 g/l stock solution; the mix was stirred using a magnetic stirrer at 500 rpm for 30 min before the experiment [30]. The powder is prepared one day before each experiment to avoid any change in its characteristics [31].

Aluminum sulphate water solution $Al_2(SO_4)_3 \cdot 18H_2O$ was used with lab-grade purity. The alum solution is prepared by dissolving the amount of Alum in distilled water.

3.2 Sampling site

Water samples were collected in November 2024 from the Tigris River, Baghdad, near Al Ahrar bridge, with the following coordinates: 33°20′00.71″ N, 44°23′43.13″ E.

Table 1 shows the results of raw water characteristics.

Table 1 – Raw water characteristics

Parameter	Mean ± st. deviation	WHO* standards
Temperature, °C	19.2	n/a**
pH	6.6 ± 0.01	6.5 – 8.5
Turbidity, NTU	76.3 ± 1.5	≤ 5
TDS, mg/l	255 ± 1.0	≤ 1000
COD, mg/l	9 ± 1.1	≤ (10–20)
Electrical conductivity, μS/cm	591 ± 1.5	≤ 1000
Dissolved oxygen, mg/l	7.5 ± 0.14	≥ 5.0

* according to the World Health Organization (WHO);
** not applicable.

Like other rivers in urban areas, this river serves as the primary recipient of industrial and municipal wastewater and runoff from agricultural lands, streets, and manure discharges, contributing significantly to river pollution.

According to a previous National Sanitation Foundation Water Quality Index (NSFWQI) study, the water quality was fair and unsuitable for human use [32].

Three grab samples were collected at suitable distance from river bank, time interval between each sample is 30 min then samples were mixed to form composite sample, sampling plastic bottles were rinsed by river water 3 times then submerged under water surface about 0.3 m to collect raw water without debris and floating objects, samples were labeled and stored at 4 °C then transported to the lab, all samples were analyzed within less than 3 h.

3.3 Experimental procedure

MO was investigated as a coagulant in water treatment using a jar test apparatus, where a certain amount of Moringa stock solution was added to the apparatus beakers and the dose, pH, time, and stirring speed [33]. Turbidity and COD reduction efficiency were obtained according to the operating parameters.

Turbidity was measured using a Lovibond MD 600 photometer; a pH meter, Lovibond SD305 pH/ORP, was used to read pH. COD was determined using a Lovibond

RD-125 thermoreactor and MD-200 photometer, and electrical conductivity was determined using Lovibond SD-320.

All the samplings and readings were conducted based on the standard methods for examining water and wastewater [34].

In coagulation experiments, the jar test apparatus is used extensively. Different amounts of natural coagulant were added to the jar test beakers. The study changed the water acidity by 0.1 M of NaOH and HCl [35]. Different coagulant dosages were added (50 to 300 mg/l) to investigate the optimum coagulant dosage, and then the optimum dosage was used at various pH values.

After adding the water sample to the beaker, the desired coagulant was added and mixed at a speed of 150 rpm for two minutes, then at 30 rpm for 20 minutes. Further settling was allowed for 30 min while the mixer was off.

Figure 2 shows the jar test before and after treatment.

Figure 2 –Jar test at MO dose (50, 100, 150, 200, and 300 mg/l)

A triplicate procedure was adopted to ensure the reproducibility and reliability of results; three samples were collected carefully from the beaker's center and tested for turbidity [36]. The percent of turbidity removal was calculated according to the following equation:

$$\%\, Turbidity\ removal = \frac{T_1 - T_2}{T_1} \cdot 100\ \%, \qquad (1)$$

where T_1 – the turbidity of the raw sample (control sample); T_2 – the turbidity after treatment.

For COD measurement, 2 ml of sample water were added to the kit vials of Lovibond low range (3–150 mg/l), close the vial and invert it several times to mix the contents the put it in preheated thermo-reactor for 120 min at 150 °C, remove the vial from the thermoreactor, allow vial to cool to 60 °C, mix the contents by inverting the vial, allow it to cool to the room temperature then take the reading with the photometer. The above procedure uses 2 ml of distilled water for a control reading [37, 38].

The percent of COD removal was calculated according to equation (2):

$$COD\ Removal = \frac{COD_1 - COD_2}{COD_1} \cdot 100\ \%, \qquad (2)$$

where COD_1 – the COD before treatment; COD_2 – the COD of the sample after treatment.

SigmaPlot 15.0 software was used for data analysis.

Table 2 illustrates the instruments used in this study.

Table 2 – Instruments used in the study

Parameter	Equipment
Hydrogen potential	Lovibond SD305 pH/ORP
Turbidity, NTU	Lovibond MD 610 photometer
COD, mg/l	Lovibond RD-125 thermoreactor; Lovibond MD-200 photometer
Electrical conductivity, µS/cm	Lovibond SD-320
TDS, mg/l	Lovibond SD 325
Dissolved oxygen, mg/l	Lovibond SD 400 Oxi-L

4 Results

The relation between turbidity removal efficiency and coagulant dose (MO and Alum) is presented in Figure 3.

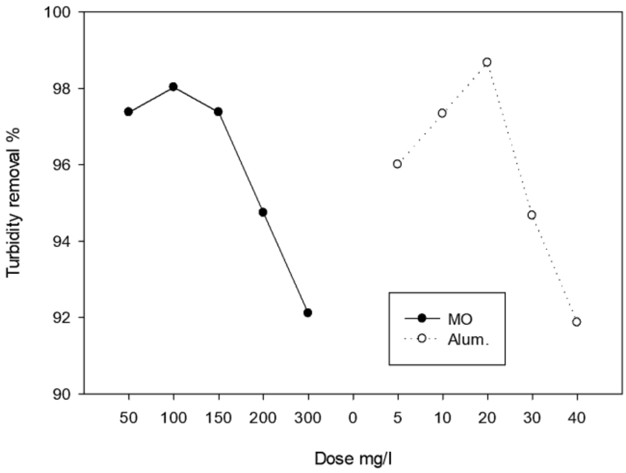

Figure 3 –Relation between turbidity removal efficiency and coagulant dose (MO and Alum)

Figure 3 shows that increasing the MO dosage from 50 mg/l to 100 mg/l, the turbidity removal rose to 97.3 ± 0.08 %, so it may be considered the optimum dose. By increasing MO concentration in the solution, a decrease in the removal efficiency was noticed because the dosage is more than the amount of pollutants in the water [39].

Functional groups on the amino acids (arginine, lysine, and histidine) are responsible for the coagulation activity [40, 41]. These findings were in line with outcomes obtained by previous research that shows water turbidity is reduced after treatment with MO [42, 43].

By comparison with the coagulation using Alum, we can conclude that a lower dosage of Alum (20 mg/l) achieved higher turbidity removal at 98.4 ± 0.25 %, and a higher Alum dose also decreases the turbidity removal efficiency.

Figure 4 shows the difference between raw and treated water using an MO dose of 100 mg/l. The relation between MO dose and the COD removal was examined as shown in Figure 5.

COD removal is increased by increasing the dose until 150 mg/l; further increases in the MO dose cause an increase in the organic matter in the water, which is one of the disadvantages of using natural coagulants.

The MO is reported as a non-toxic material used for water purification [44, 45].

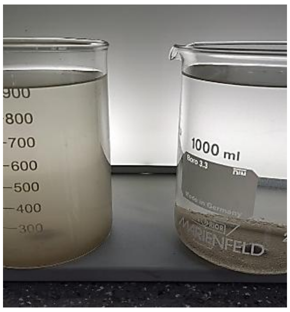

Figure 4 – Difference between raw water and treated water at optimum MO dose (100 mg/l)

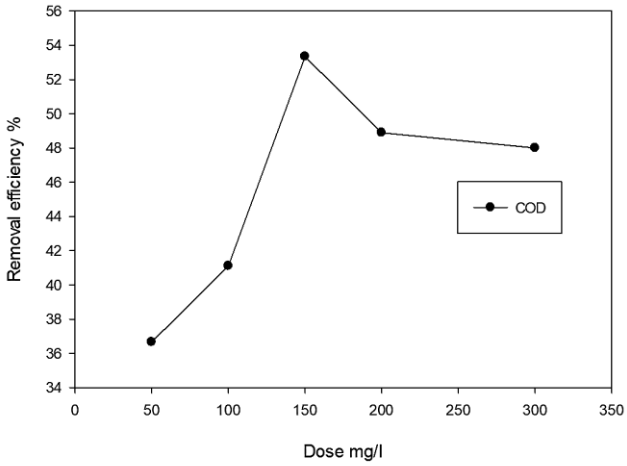

Figure 5 – Impact of MO dose on COD removal efficiency

Figure 6 shows the relation between the dose and the pH, at a dose of 50 mg/l; the pH reading was 6.55 ± 0.005.

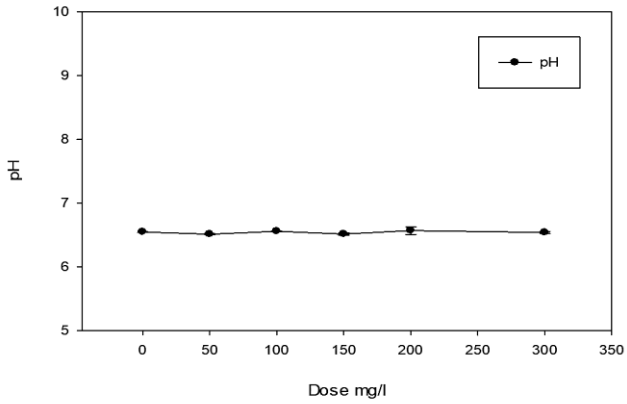

Figure 6 – Effect of MO dose on pH

When the dose was raised to 300 mg/l, the pH reading was 6.53 ± 0.01. MO dosing had no significant effect on changing the solution's pH; the same findings were concluded by [41, 46].

The effect of dose on electrical conductivity was studied as shown in Figure 7. It can be concluded that increasing the dose will increase the electrical conductivity (EC) from 591 ± 1.5 μS/cm to 620 ± 1.5 μS/cm ($P \leq 0.05$). The results found were similar to those obtained by [43].

The same conclusion was adopted for the dosage effect on the TDS as represented in Figure 8.

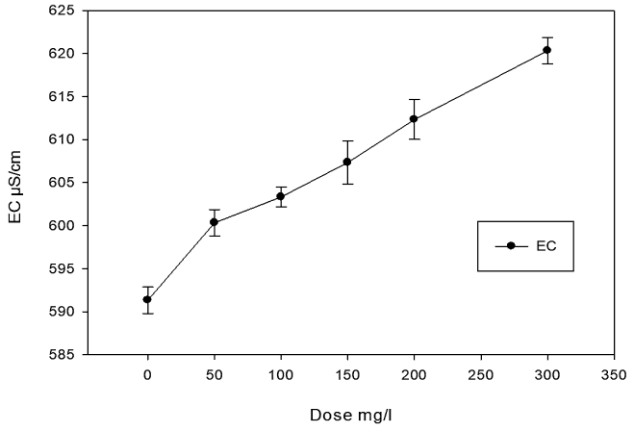

Figure 7 – Effect of MO dose on electrical conductivity

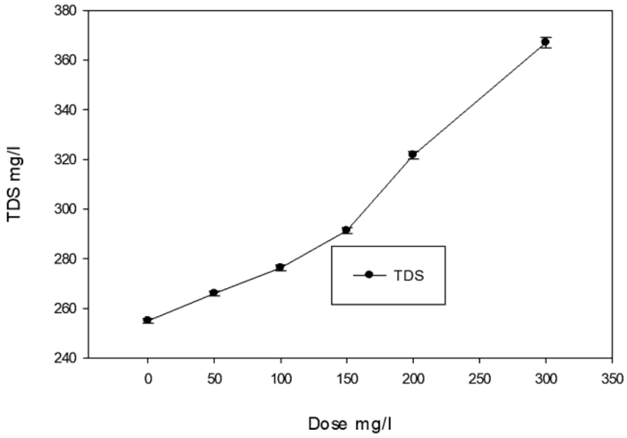

Figure 8 – Dosage effect of MO on the TDS

Turbidity removal was studied using different pH values. Figure 9 shows that the removal increased in basic solution due to the increase of the negative charge on the coagulant surface, while the higher hydrogen ions in the acidic solution will neutralize the negative charge of the coagulant surface, leading to a reduction in the adsorption mechanism.

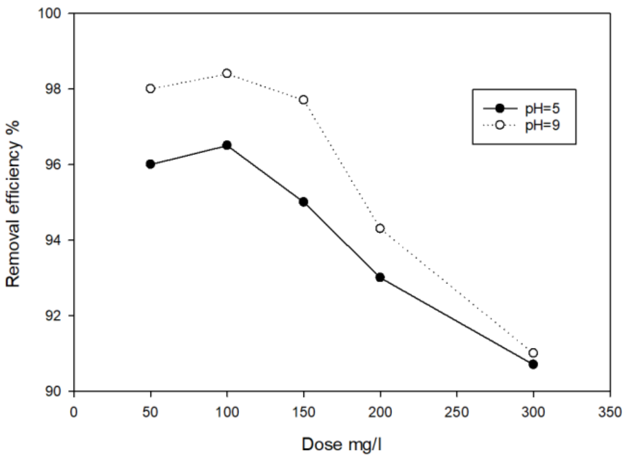

Figure 9 – Effect of MO dose and pH on turbidity removal

The relation between dose and turbidity removal was examined statistically using the analysis of variance (ANOVA) method.

Table 3 shows that the regression model is significant (F-value of 76.1 and P-value less than 0.0001).

Table 3 – ANOVA test

DoF	Sum of squares	Mean square	F-value	P-value
2	75.1	37.55	76.1	< 0.0001
12	5.92	0.494		
14	81.0	5.787		

* degrees of freedom (DoF).

These values suggest that the results from the statistical test indicate a strong effect, and the evidence against the null hypothesis is very compelling.

The coefficient of determination R^2 of 0.93 suggests the model is well-fitted to the data and explains nearly the entire variability in the dependent variable. All the data collected were from experiments with low turbid water (not more than 100 NTU), so the application of this model in the following equation is applicable for this turbidity range:

$$y(x) = 97.2847 + 0.0134 \cdot x - 0.0001 \cdot x^2, \qquad (3)$$

where x – the dose, mg/l; y – the turbidity removal, %.

Figure 10 shows the correlation between the experimental and predicted values.

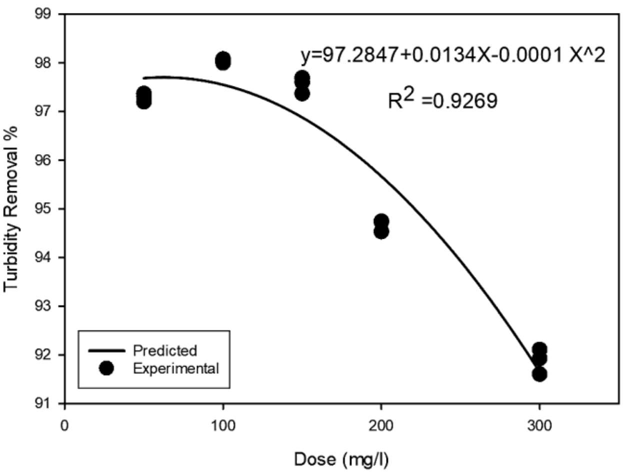

Figure 10 – Correlation between experimental and predicted

The second-order polynomial equation (3) shows a nonlinear behavior in which turbidity removal efficiency increases with increasing MO dose but yields an optimum dose after which efficiency decreases.

5 Discussion

A mild increase was noticed in turbidity removal efficiency by increasing the MO dose to 100 mg/l and the Alum dose to 20 mg/l, as shown in Figure 3. Alum and MO seed powder showed promising results for turbidity removal (98.4 % and 97.5 % respectively).

However, Alum had higher efficiency than MO due to its highly soluble purified chemical works through charge neutralization, while MO contains active proteins in low concentration and acts through adsorption and interparticle bridging [47].

Initially, increasing coagulant dosage led to enhanced turbidity removal efficiency, reaching an optimal dosage level at 100 mg/l of MO. However, increasing MO dosage beyond the optimal dosage resulted in a marked decline in turbidity removal performance. Similar trends can be noticed when using Alum as a coagulant. This trend is attributed to the fact that at sub-optimal dosage, the number of cations is insufficient to neutralize the negative surface charges of colloids, meaning that repulsive electrostatic forces are high. At optimal dosage, the zeta potential is expected to be around zero, meaning that charge neutralization is almost complete, resulting in large flocs that settle rapidly and peak removal efficiency. On the other hand, overdosing may have a floc structure, reducing the settling and overall removal efficiency. It may also indicate an excess of cations compared to anions [48].

The relation between MO dose and COD was illustrated in Figure 5. COD removal followed a similar pattern to turbidity, where removal efficiency declined beyond optimum dosage. At the optimal MO dose, organic matter with a negative charge is also eliminated by a sweep coagulation mechanism.

Compared to turbidity removal efficiency, a slightly higher MO dose is required for COD removal because COD may be attributed to a wide range of dissolved stable organics. However, when all the charges are neutralized, increasing the coagulant dosage may change the charge of the colloidal substances, releasing again adsorbed organic matter, which reduces COD removal efficiency; thus, finding an optimum dosage is essential in coagulation.

Different MO dosages did not cause a significant change in pH values, as illustrated in Figure 6. The effectiveness of MO in the coagulation process is due to the existence of water-soluble cationic proteins, which are weakly acidic or neutral [49]. It is suggested that basic amino acids in the protein molecules have a minor ability to receive a proton from a water molecule, leading to the release of a hydroxyl group, so the addition of MO to the water will not affect the pH significantly [50].

In Figure 7, electrical conductivity increased by increasing MO dosage, due to the dissociation of coagulant producing ions [51] and the increase in cationic polyelectrolyte in MO, generating high dissolved solids that raise the conductivity [52]. Simultaneously, TDS increment is caused by different organic compounds in the seeds, including coagulation-active and inactive components [53, 54].

The quadratic model fits the experimental data collected in the 50–300 mg/l dosage range, with an R^2 of 0.93, indicating a strong correlation. It could be used in estimating turbidity removal without extensive experimentation, identifying optimal MO dose, noting that this empirical model is specific to the tested conditions, including initial turbidity level (60–90 NTU), raw water characteristics, and mixing regimes.

6 Conclusions

Moringa oleifera (MO) plant has a rapid growth rate; unskilled workers can easily produce and use its extract without the hazard of dealing with chemical compounds. So, the coagulant extracted from MO may be available in large quantities.

Natural coagulants in water treatments have been investigated and compared to using aluminum sulfate (Alum) in coagulation, and relations between MO and electrical conductivity (EC), pH, and total dissolved solids (TDS) were studied. The effect of different pH values on MO in turbidity removal was also investigated.

The optimal MO dose for turbidity removal was 100 mg/l to achieve 97 % removal, while for Alum, it was 20 mg/l to reach about 98 %. For COD, the highest removal was about 49 % at 150 mg/l MO dosage. The results significantly enhance the effectiveness of MO as a good alternative to chemical coagulants in the water treatment process.

TDS also increased for raw water from 255 ± 1 to 367 ± 2 at 300 mg/l. Increasing the dose will increase the EC from 591 ± 1.5 μS/cm to 620 ± 1.5 μS/cm. An optimal MO dose as a coagulant depends on the initial water characteristics. So, the turbidity reduction obtained in this study confirms the significant eco-friendly natural material possibilities in the water treatment process.

It could be recommended that further research on extracting and isolating the active components economically from MO powder that are responsible for the coagulation mechanism, study the mixing of two or more coagulants (natural or artificial), and evaluate this natural coagulant in heavy metal removal from water. Also, the characteristics and management of the sludge produced after the coagulation process will be studied to assess the MO environmental impact for safe disposal and reuse.

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