

## Assessing the efficacy of bentonite as a coagulant aid for raw water treatment

Najlae Zaki<sup>a</sup>, Nouhaila Hadoudi<sup>a</sup>, Nihade Bensitel<sup>b</sup>, Amin Salhi<sup>a</sup>, Hassan Amhamdi<sup>a</sup>, M'hamed Ahari<sup>a,\*</sup>

<sup>a</sup>Applied Chemistry Team, FSTH, Abdelmalek Essaâdi University, Tetouan, Morocco, Tel.: +212637568299; Fax: +212539807173; emails: najlaezaki5@gmail.com (N. Zaki), nouhailahadoudi79@gmail.com (N. Hadoudi), a.salhi@uae.ac.ma (A. Salhi), h.amhamdi@uae.ac.ma (H. Amhamdi), m.ahari@uae.ac.ma (M. Ahari)

<sup>b</sup>Laboratory of ONEE-Water Branch, Treatment Station, Al Hoceima, Morocco, email: nihade.bensitel@etu.uae.ac.ma

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### ABSTRACT

This study aims to demonstrate the efficacy of both raw and pure bentonite as a coagulant/adjuvant in treating raw water from the Al Hoceima plant. The focus of this study lies in the evaluation of two distinct coagulant combinations with bentonite: aluminum sulfate/bentonite and chitosan/bentonite. Additionally, an assessment is made on the contrasting efficiencies of pure and raw bentonite in coagulation–flocculation processes. The potential link between Alzheimer's disease and aluminum has raised concerns due to the presence of aluminum residues after the treatment of raw water. By employing a natural organic coagulant, it becomes possible to mitigate environmental and human health risks. The experimentation involved various sections of jar tests. These tests encompassed the addition of sole coagulants (either aluminum sulfate or chitosan) as well as tests utilizing coagulant/adjuvant combinations (either aluminum sulfate or chitosan/bentonite) for raw water treatment. Key parameters under scrutiny included turbidity, pH, aluminum, and oxidizability. Results underscore that the inclusion of 30 mg/L of bentonite as an adjuvant alongside aluminum sulfate led to a 95.11% reduction in turbidity and a 56.75% decrease in oxidizable content. Similarly, the incorporation of 250 mg/L of bentonite with chitosan resulted in a 95.26% decrease in turbidity and a 57.39% reduction in oxidizable matter. Moreover, a comparative analysis between raw and pure bentonite in conjunction with coagulants was performed. Bentonite exhibited a discernible impact on the coagulation/flocculation process, contributing to larger floc formation, enhanced sedimentation rates, and influencing pH levels.

*Keywords:* Aluminum sulfate; Bentonite; Chitosan; Coagulation/flocculation; Oxidizability; Turbidity

### 1. Introduction

Water is considered an essential element. This resource presented in dams in large quantities. The presence of contaminants in dams creates problems in raw water treatment. Many techniques exist to eliminate these pollutants, suitable for raw water quality to make it drinking water [1]. Among the most effective and important techniques are the coagulation/flocculation methods [2,3], which are the simplest and most cost-effective ways to remove suspended

matter [4]. The correct application of the coagulation process and choice of coagulant depends on understanding the interaction between different factors [5,6]. Most of the stations use chemical coagulants, aluminum salts, or iron salts. The most commonly used in Morocco are aluminum sulfate and ferric chloride which are added during treatment [7]. Several epidemiological studies have suggested a potential link between aluminum ions in water and Alzheimer's disease [8]. These studies have investigated the hypothesis that high levels of aluminum exposure through drinking water

\* Corresponding author.

might be associated with an increased risk of developing the disease. However, the use of a natural polymer (chitosan) as a coagulant has proven effective in different studies [9], chitosan extracted from waste shells of crustaceans or fungal biomass [10]. Chitosan is soluble in solutions with a pH inferior to 6.2 [11,12]. The addition of chitosan provides an opposite ionic charge to the colloidal particles in the wastewater, neutralizing the charges [13]. Many researchers were inspired to study the clay/coagulant combination [14,15]. Using bio-adsorbents as adjuvants like bentonite mineral clay allows for a decrease in the coagulant dose and gives more efficiency to the process.

The main goals of this study are to evaluate the effect of local bentonite as a coagulation aid combined with the coagulants aluminum sulfate and chitosan (inorganic and organic coagulants) in water treatment. Moreover, to study the effectiveness of (raw and pure) bentonite in removing turbidity and oxidizable materials from raw water in the production of Al Hoceima drinking water.

## 2. Materials and methods

### 2.1. Location of the water production station of Al Hoceima

Built-in 1975 and launched in 1985, the drinking water production plant of Bni Bouayach is one of the departments of ONEE – Water Branch and supplies all the needs of the Al Hoceima population region in drinking water (Fig. 1), with a flow that can reach up to 440 L/s, the supply being provided by the Mohammed Ben-Abdelkrim Khattabi (Fig. 2). It was built on Oued Nekkour, and had a capacity of 50 Mm<sup>3</sup> at the beginning of construction which was decreased to 116 Mm<sup>3</sup> in 2019 by a large number of solid matter inputs. Oued Ghiss is another source that supplies drinking water to Al-Hoceima region.

### 2.2. Physicochemical analysis

A raw water sample is collected from the distribution structure at the station. Several parameters are analysed to

get an overview of this water quality. Table 1 summarizes the various parameters, techniques and method used.

### 2.3. Sampling

The clay samples used for these analyses were collected from Nador located in the North-East of Morocco [16,17]. The clay used is sodium bentonite, gray color [14–18]. First, a series of washing to remove impurities after clay samples were subjected to dry, crushing, and sieving operations at the laboratory [10]. The clay materials were sieved through 0.63 mm. It is composed principally of montmorillonite [15]. A stock suspension of bentonite (raw and pure), chitosan, and aluminum sulfate at a concentration of 10 g/L is prepared by dispersing in distilled water. The chlorine stock solution is prepared by dissolving chlorine bleach in distilled water.

The clay used is bentonite to purify this clay we start with a successive washing with distilled water, sedimentation allows to remove of impurities and large particles, the supernatant part is separated by centrifugation and then dried in the oven at 60°C for 24 h and crushed until a



Fig. 2. Mohammed Ben-Abdelkrim Khattabi dam.



Fig. 1. Location of drinking water production station.

homogeneous powder is obtained. The resulting product was washed with H<sub>2</sub>O<sub>2</sub> to oxidize the organic matter. The sample was then washed (three times) with NaCl and centrifuged for 24 h, the grey part of the product which is in the centrifugation tube was eliminated due to its enrichment by impurities after the clay was washed with distilled water. The mineral part was recovered from size <2 μm by sedimentation then dried in the oven at 80°C and ground to obtain a powder.

2.4. Coagulation/flocculation (jar test)

A Series of coagulation/flocculation tests were carried out using jar-test JLT6 of 1 L beakers of raw water [15], with a rapid agitation of 120 rpm for 2 min followed by flocculation of 40 rpm for 20 min. The flocculated waters were precipitated for 30 min. During rapid agitation doses of coagulant, adjuvant, and chlorine additive have been added (3 mL of chlorine has been added to disinfect the water). Four sections of the tests were used to determine the optimal dose of coagulants and adjuvant. First section treatment tests were carried out by adding increasing doses from 10 to 45 mg/L of aluminum sulfate coagulant alone in the beakers. Secondly, section was performed by adding increasing doses of chitosan alone from 10 to 100 mg/L in the beakers. The two other sections, doses of coagulants and adjuvant were added to the beakers. For the tests of aluminum sulfate and bentonite, the dose of bentonite was fixed (10–20–30–40 mg/L) in each test with a variation of the dose of aluminum sulfate coagulant from 10 to 120 mg/L. In the section of adding chitosan and bentonite, the chitosan dose was fixed each trial (10–20–40 mg/L) and the bentonite dose was increased (250–500–750 mg/L).

Furthermore, next step is to find the optimal dose of coagulant and adjuvant from the measurement of the parameters and choose the values suitable for drinking water standards.

3. Results and discussions

3.1. Characterization of the raw water of the station of Al Hoceima and bentonite

The drinking water station of Al Hoceima is supplied by the dam of Mohammed Ben-Abdelkrim Khattabi dam.

The physico-chemical parameters were analysed according to the method standard AFNOR 1999 (French National Organization for Standardization) [19]. An analysis of the results which are presented in Table 2 indicates that the alkalinity (TAC) is bicarbonate, as the pH is less than 8.3. However, TAC is related to chlorides and sulfates. The high sulfate values are caused by the geological nature of the dam soil and the parameters of the raw water are constant. The scanning electron micrographs of bentonite composites that were taken at 3,000 magnifications are shown in Fig. 3. While the image highlights the near homogeneity of the nanocomposite, it also distinctly portrays the heterogeneous structure of the bentonite’s surface. This structural heterogeneity is a characteristic feature of highly porous materials, suggesting that the biosorbent has the potential to adsorb pollutants across diverse regions [20].

3.2. Effect of aluminum sulfate alone

The jar test was based on this part by adding aluminum sulfate only, in order to determine the optimal dose of aluminum sulfate, coagulant doses were introduced between 10–45 mg/L to determine the optimal dose. Upon the addition of alum under rapid agitation, hydrolysis reactions ensue, leading to the creation of four principal dissolved monomers. These hydrolysis reactions yield four primary dissolved monomers: Al<sup>3+</sup>, Al(OH)<sup>2+</sup>, Al(OH)<sub>2</sub><sup>+</sup>,

Table 2 Results of physico-chemical analysis of the raw water of the plant

Parameter	Min. – Max.	Average
Temperature, °C	14.9–16	15.5
pH	8.06–8.23	8.15
Turbidity, NTU	24.2–26.5	25.40
Conductivity, μS/cm	3,120–3,140	3,130
TAC, meq/L	3–3.20	3.10
Chloride, mg/L	674.5–727.75	701
Sulfate, mg/L	979.91–1,029.79	1,004.85
Oxidizability, (mg-O <sub>2</sub> /L)	4.2–4.5	4.35
Hardness, mg/L	21.6–23.6	22.6

Table 1 Techniques and methods used for the chemical characterization of raw water parameters

Parameter	Method and technique
Turbidity	Nephelometry (turbidity meters Turb 300 IR)
pH	Electrometric measurement (HQ40d Dual-Input Multiparameter)
Conductivity	Electrical conductivity (HQ40d Dual-Input Multiparameter)
TAC	Determination by hydrochloric acid HCl (with helianthin), titrimetric method
Chloride	Determination by mercuric nitrate, Mohr method
Sulfate	Determination by hydrochloric acid HCl (with barium chloride), (turbidity meters)
Oxidizability	Hot acid oxidation by KMnO <sub>4</sub>
Hardness	Determination by ethylenediaminetetraacetate
Aluminum	Aluminum analysis kit with visual colorimetric comparator

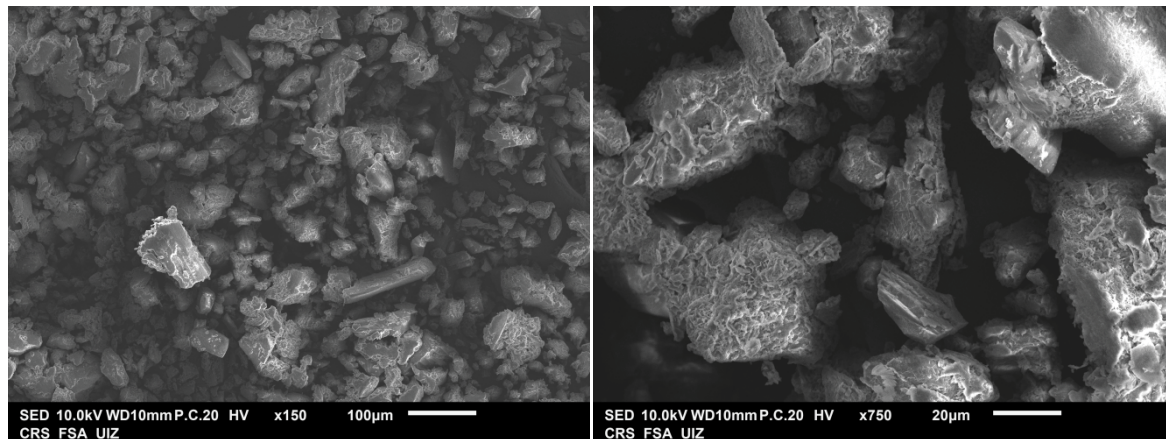


Fig. 3. Scanning electron micrographs of raw bentonite composite [20].

and  $\text{Al}(\text{OH})_4^-$ . Based on the reaction kinetics between various Al species and ferrous reagents, hydrolysed Al species can be categorized into three distinct types: Al monomers, Al polymers, and colloidal Al [21,22]. From the results obtained in Fig. 4, pH decreases by increasing the dose of aluminum sulfate but the doses 40–45 mg/L, pH increase due to excess aluminum sulfate. The efficiency of charge neutralization coagulation is primarily influenced by the zeta potential of both flocs and the precipitates formed by coagulants [23]. Reducing the zeta potential and charge can be achieved through the introduction of flocculants carrying an opposing charge. This facilitates an improved collision efficiency among colloidal particles suspended in the suspension. Following an increase in a specific concentration of bentonite, the zeta potential either remained relatively stable or exhibited a slight decrease [24].

For the parameters turbidity and oxidizability (Fig. 5) decrease by increasing the dose of aluminum sulfate. The aspect of flocs increases as the coagulant dosage increases, as indicated in Table 3, while the suspended solids concentration in raw water is 0.1 mg/L. The optimal dose of aluminum sulfate for drinking water standards is 35 mg/L.

### 3.3. Effect of using bentonite and aluminum sulfate

Jar tests were based on adding coagulant and adjuvant sequentially. The hydrolysed  $\text{Al}^{3+}$  ions, carrying a positive charge, are attracted to the negatively charged colloidal particles of bentonite. This electrostatic attraction promotes the formation of complexes between these  $\text{Al}^{3+}$  ions and the colloidal particles, thereby creating aggregates known as flocs. These flocs, composed of clustered particles, can also trap other suspended particles in the water. The hydrolysed  $\text{Al}^{3+}$  ions partially neutralize the negative charges on the bentonite particles, thus reducing their electrostatic repulsion [22]. This action facilitates the approach of particles and encourages their coagulation. The flocs continue to form and sediment under the influence of gravity. The parameters were measured after the beakers were decanted.

The graphs generated from coagulation tests using the coagulant/adjuvant combination illustrate how the different

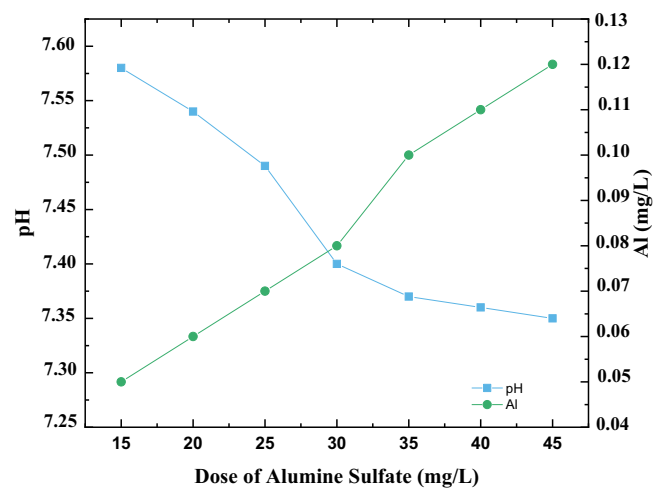


Fig. 4. Variation of pH and aluminum as function of aluminum sulfate.

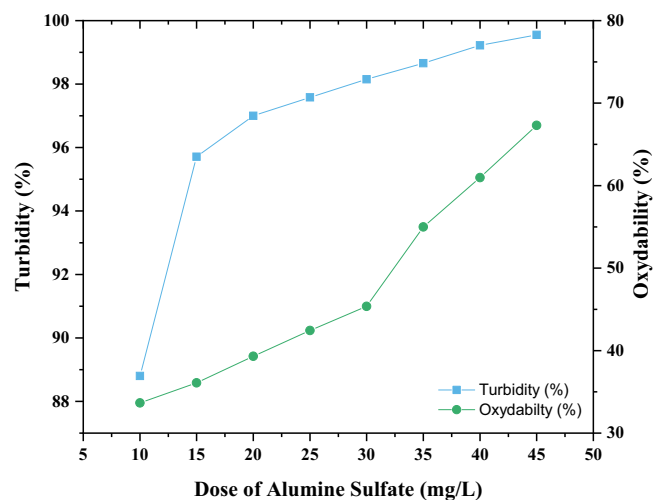


Fig. 5. Variation of turbidity and oxidizability as function of aluminum sulfate.

Table 3  
Results of the coagulation/flocculation test using aluminum sulfate

	Injected dose of aluminum sulfate (mg/L)							
	10	15	20	25	30	35	40	45
Aspect flocs	04	06	06	06	08	08	08	08
Settling speed	Slow (S)	S	Medium (M)	M	Rapid (R)	R	R	R

\*Aspect flocs: 4 = Small flocs, 6 = flocks of medium size, 8 = large flocks.

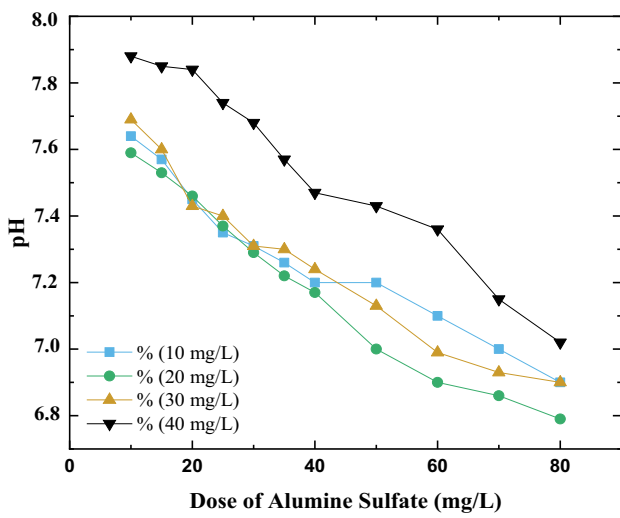


Fig. 6. Variation of pH with SA dose.

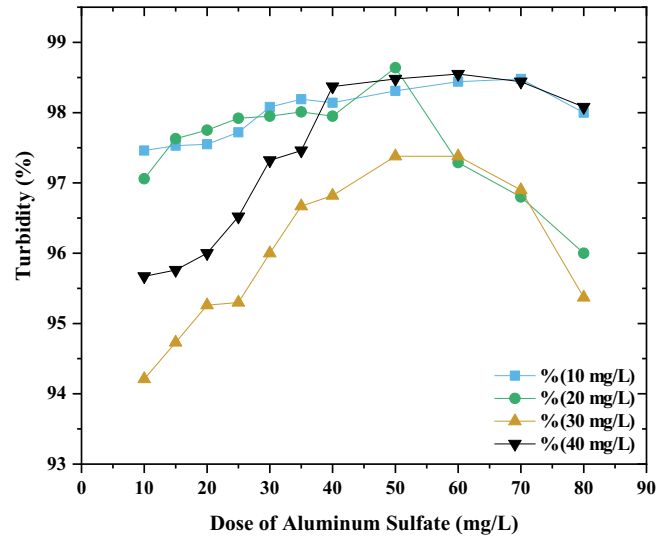


Fig. 7. Variation of turbidity removal rate with SA dose.

dosages of coagulant and bentonite impact the variations in parameters such as pH, aluminum concentration, aspect flocs, turbidity, and oxidizability. The observation reveals that elevating the dosage of aluminum sulfate (SA) leads to a reduction in pH (Fig. 6), turbidity (Fig. 7), and oxidizability (Fig. 8). This decrease can be attributed to the increase in aluminum sulfate concentration. This effect finds an explanation in the phenomenon of organic matter adsorption onto the flocs formed during the process. The variation of pH is affected by the variation of organic matter present in the water. Aluminum increases with increasing dose of aluminum sulfate (Fig. 9). The addition of 70–80 mg/L of aluminum sulfate decrease turbidity, this increase is due to the excess of aluminum sulfate. Mineral clays can be used as adjuvant with primary coagulants such as aluminum sulfate in the coagulation step to bind the small flocs already formed into larger particles [25,26]. The size of the flocs depends on the dose of aluminum sulfate injected, increasing the dose of aluminum sulfate increases the size of the flocs which facilitates their settling (for 10 and 15 mg/L the size of the flocs was small for the other doses the flocs were large). Further research has discovered that there are specific values for velocity gradient and mixing time values that can be considered optimal [27]. These values work towards minimizing the remaining turbidity within a defined set of conditions. The optimal dose found based on the parameters is 30 mg/L of bentonite combined with 25 mg/L of aluminum sulfate which was able to remove 95.11% of turbidity and 56.75% of oxidizable matter.

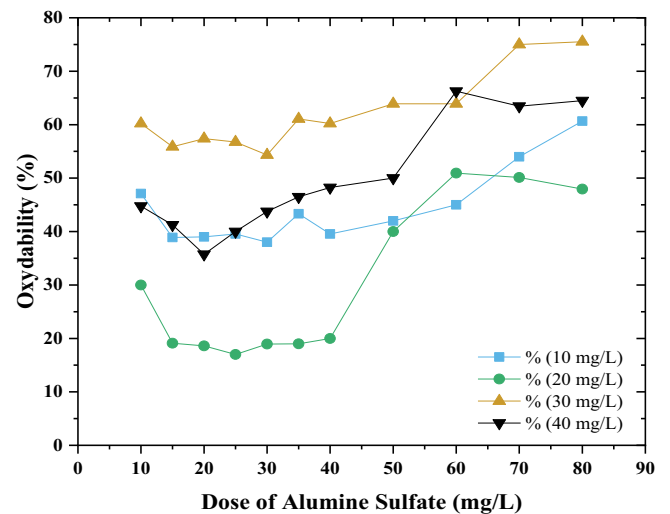


Fig. 8. Variation of oxidizable matter removal rate with SA dose.

### 3.4. Effect of using chitosan alone

In this series of experiments carried out, chitosan is used as a coagulant. To determine the optimal dose of chitosan, coagulant dose was introduced between 10–100 mg/L. The parameters were measured after the coagulation/flocculation–decantation processes.



According to the results presented in Table 4, chitosan shows that hardly can have effective results on the removal of suspended particles in water. At a higher pH, the positive charges of chitosan can decrease due to the deprotonation of the amino functional groups (NH<sub>2</sub>), which can diminish its ability to attract and aggregate negatively charged suspended particles. The effect of pH on the efficiency of chitosan can depend on several factors, including the concentration of chitosan, the nature of suspended particles, and other components present in the water [28].

3.5. Effect of using chitosan and bentonite

This section presents 3 trials to study the role of the adjuvant bentonite (BN) in combination with chitosan. According to the results presented in Figs. 9–11, the assembly of bentonite which is an ideal colloidal particle combined with chitosan increased the efficiency of the coagulation/flocculation process and the rate of turbidity according to the time until reaching 95.26% of removal turbidity and 57.39% of oxidizability rate. During the treatment, the different charges of two substances combine (negative charge due to bentonite and positive charge due to chitosan). This attraction promotes the aggregation of both bentonite particles and chitosan molecules. Only minimal quantities of chitosan were necessary to induce the destabilization and

settling of concentrated bentonite suspensions, leading to extremely low turbidity levels attained within a brief settling period of just a few minutes [29]. The sedimentation

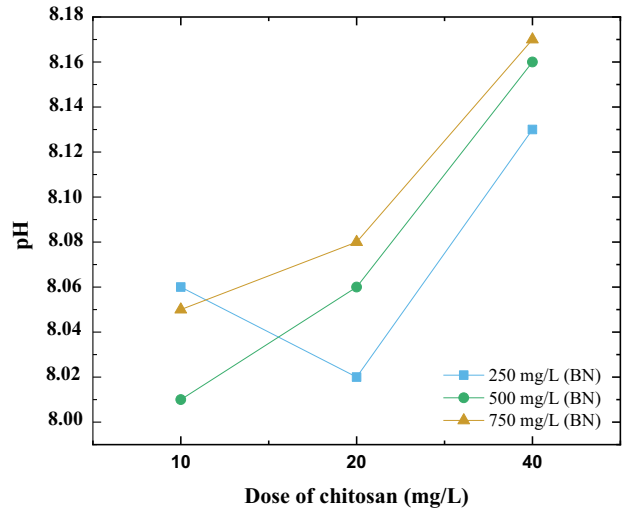


Fig. 9. Variation of pH as a function of bentonite and chitosan.

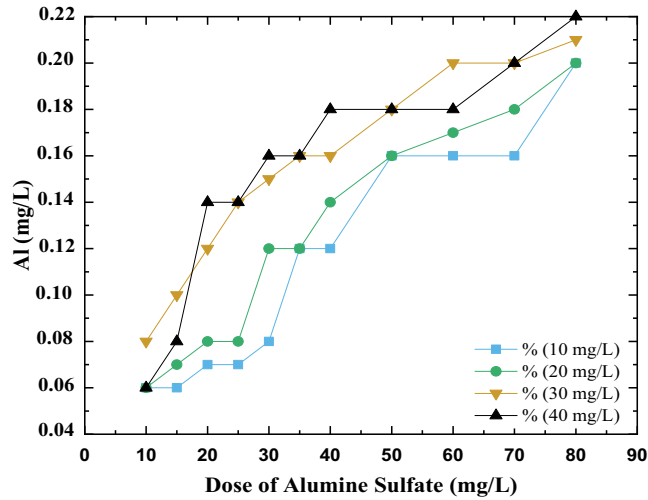


Fig. 9. Variation of aluminum with SA dose.

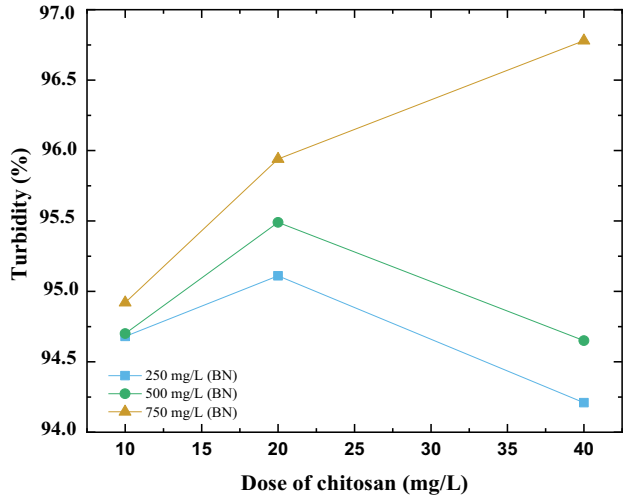


Fig. 10. Variation in the rate of turbidity as a function of bentonite and chitosan.

Table 4 Results of using chitosan alone

Dose of chitosan (mg/L)	Physico-chemical parameters							
	pH	Turbidity		Tur. rate	Aspect flocs	Settling speed	Oxidizability (mg·O <sub>2</sub> /L)	Ox. rate
		D.W. (NTU)	F.W. (NTU)					
10	7.99	32.1	10.8	14.4	04	S	1.8	28
40	7.92	26.0	10.3	30.67	04	S	1.3	48
60	7.97	25.7	7.69	31.47	06	M	1.64	34.4
80	7.95	27.0	9.78	28	08	R	1.46	41.6
100	7.96	26.3	9.82	29.87	08	R	2.46	1.6

Table 5  
Comparison of studies between aluminum sulfate/bentonite and chitosan/bentonite

No.	Coagulant/adjuvant	pH	1st run/2nd run (rpm)	Optimum dose of coagulant/adjuvant (mg/L)	Elimination rate	References
1	Sulfate aluminum/bentonite	7.40	120 (2 min)/40 (20 min)	25/30 mg/L	95.11% of removal turbidity and 56.75% oxidizability rate	This study
2	Sulfate aluminum/bentonite	7.26	120 (2 min)/40 (20 min)	50/20 mg/L	96.72% of removal turbidity and 60% of the oxidizable material	[15]
3	Sulfate aluminum/bentonite	–	200 (1 min)/50 (20 min)	10/10 mg/L	88.92% of removal turbidity	[34]
4	Chitosan/bentonite	8.02	120 (2 min)/40 (20 min)	20/250 mg/L	95.26% of turbidity and 57.39% of oxidizability	This study
5	Chitosan/bentonite	5	100 (2 min)/30 (30 min)	1,000 mg/L (with 0.15 g chitosan + 0.35 g bentonite)	98% of turbidity removal	[32]
6	Chitosan/bentonite	7.90	100 (2 min)/30 (30 min)	1,000 mg/L (300 mg: 700 mg in 1 L of raw water)	97.23% of turbidity removal	[35]

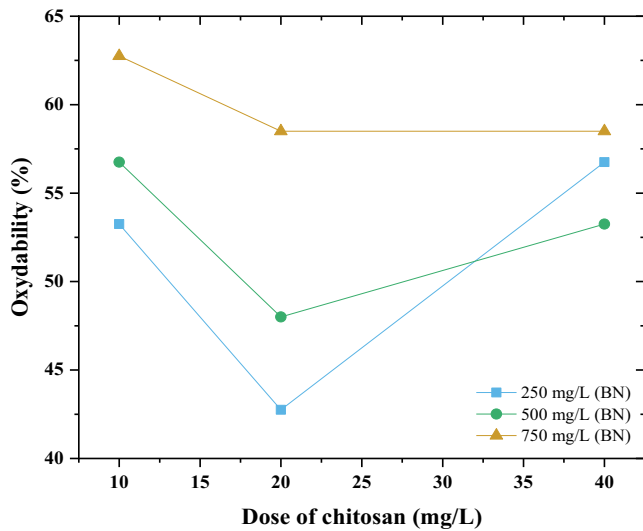


Fig. 11. Variation in the rate of elimination oxidizability as a function of bentonite and chitosan.

process was rapid, which made it possible to give large flocs better than the use of chitosan alone [30]. Flocs were found around the agitator blade during the flocculation period and formed spongy, jelly-like masses called [31], which can cause difficulties when combining chitosan and coagulation aids. The quality of the water treated with bentonite and chitosan was better than using bentonite and aluminum sulfate because there was no residual aluminum in the treated water, unlike aluminum sulfate [32].

To compare the results obtained with the studies carried out in the last decades (Table 5), the optimal doses of coagulants and adjuvant are varied according to the variation of the parameters of the water to be treated: pH, turbidity.

The use of chitosan and bentonite is more effective than aluminum sulfate and bentonite. The different pH values were adjusted with H<sub>2</sub>SO<sub>4</sub> and NaOH to be suitable for each standard [33].

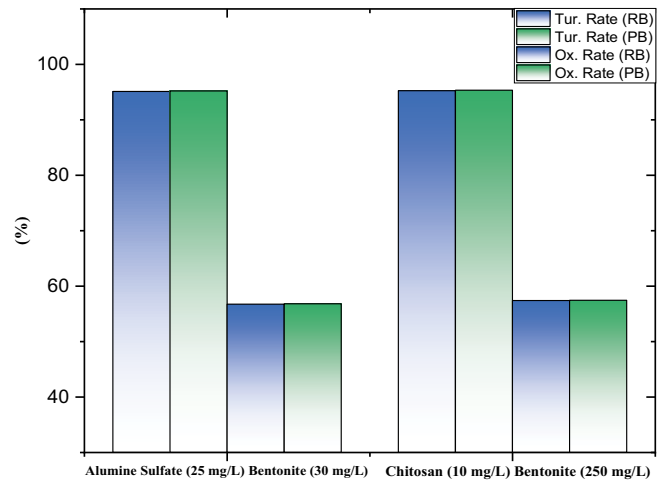


Fig. 12. Comparison between raw and pure bentonite combined with aluminum sulfate and chitosan.

### 3.6. Comparison of pure and raw bentonite

After optimal doses have been reached using raw bentonite, a study of raw bentonite (RB) and pure bentonite (PB) was compared for the following parameters: turbidity, oxidizability, floc aspect, and settling speed for the optimal doses found in the tests of aluminum sulfate/bentonite and chitosan/bentonite. The results presented in Fig. 12 show that there is not a large difference between the raw and purified clay. The flocs were large with a high settling speed.

## 4. Conclusion

The primary aim of this research was twofold. Firstly, optimize coagulant dosage (aluminum sulfate and chitosan) by synergistically employing them with bentonite. Secondly, to examine the effectiveness of bentonite (raw and pure) on the removal of turbidity and oxidizable matter in the analysed raw water.

In this study, using of organic coagulant chitosan with bentonite successfully showed their effectiveness compared to the inorganic coagulant (aluminum sulfate) with bentonite in the processes of coagulation/flocculation and settling. The use of aluminum sulfate and bentonite eliminated 95.11% of turbidity and 56.75% of oxidizable matter while using of chitosan combined with bentonite eliminated 95.26% of turbidity and 57.39% of oxidizable matter.

The use of a coagulant alone does not always give good results in the process of coagulation/flocculation like the use of chitosan alone. The addition of bentonite (adjuvant) improves raw water treatment processes. The use of bentonite with coagulants has made it possible to reduce the dose of each coagulant, the optimal dose of aluminum sulfate is 25 mg/L and for chitosan 20 mg/L, these doses have given better results in terms of the measured parameters. Using the adsorption properties of bentonite for flocculation can be an interesting solution in raw water treatment. The results indicate that the use of a coagulant inorganic (aluminum sulfate) lowers the pH, but using an organic coagulant (chitosan) increases the amount of organic matter in the water and raises the pH. Optimal pH conditions during the jar tests improved in water alkalinity results. The coagulation/flocculation process is influenced by various factors, including mixing time, initial coagulant or adjuvant addition, pH variation, and alkalinity of the raw water. In order, using chitosan and bentonite allows for an environmentally friendly situation and minimizes the risks of pathogenic diseases. This leads to an improvement in the quality of drinking water.

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### Author's contribution

The current research done by the authors.

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