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Performance of Natural Coagulant Extracted from Castanea Sativa Tree Leaves in Water Purification processes

Manar Banwan Hasan[1] | Ahmad Benwan Hassan[2]✉ | Israa M. Al-Tameemi[1] | Nawar Banwan Hassan[3]

1. Environmental Engineering Department, College of Engineering, Mustansiriyah University, Baghdad, Iraq.
2. Civil Engineering Department, College of Engineering, Al-Iraqia University, Baghdad, Iraq.
3. Imam alkadhum college (IKC), Baghdad, Iraq.

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ABSTRACT

Numerous coagulants, including natural and chemical coagulants, have been examined in the context of water purification. The use of natural coagulants constitutes an affordable and eco-friendly method of purifying water. The main aim of the current study was represented by investigated the feasibility of coagulant extracted from Castanea Sativa Tree Leaves using three different salts and distilled water. The active coagulant component was extracted using 0.25, 0.5, and 1 M of NaCl and KCl, 0.025, 0.05, and 0.1 M of NaOH, and distilled water. Powdered Castanea Sativa Tree Leaves was also used as a coagulant. Jar tests were performed using synthetic turbid water, a turbidity level of 35 NTU to investigate the coagulants' activity. The pH was measured to study the influence of a range of different pHs, coagulant doses and initial turbidity were also investigated to optimize the coagulation process. The highest level of activity was achieved using 0.5 ml/l of coagulant extracted with 0.5 M NaCl at pH level 8. Coagulant extracted using 0.05 M NaOH demonstrated the second highest level of activity. Poor coagulant activity was observed for the powdered Castanea Sativa Tree Leaves and distilled water extract. The protein content of the extracted coagulant was 0.322, 0.283, and 0.274 mg/ml using 0.05 M NaCl, 0.5 M NaOH, and 0.5 M KCl, respectively. The use of this natural coagulant was also found to moderately increase organic matter content in the treated water, which was proportional to protein contents of the extracts. Coagulation results were statistically examined using SigmaPlot 12.5 software.

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INTRODUCTION

Water shortage and pollution are global problems that can cause conflict and illness. Several kinds of contaminants and impurities can be found in water, which must undergo suitable treatment prior to being used (Abbas et al., 2020; Mohana et al., 2020; Hasan et al., 2021; Krishnaiah et al., 2015). The quality of water depends on its physical, chemical, and microbiological characteristics. Drinking water must be colorless, odorless, and free of microbes, suspended matter, and organic and inorganic contaminants that can cause illness. Suspended solids are fine particles dispersed in water known as colloids that do not tend to settle as a result of gravity (Antov et al., 2010).

Coagulation and flocculation are the standard treatment processes for turbid water. Coagulation involves the addition of coagulants to cause colloids to clump together (Cho et al., 2006). Coagulation is overwhelmingly followed by a process called flocculation (moderate

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*Corresponding Author Email: *ahmad.b.hassan@aliraqia.edu.iq*

mixing) which promotes the formation of glumes (created during coagulation) that settle as larger flocs at the bottom of the sedimentation tank due to the gravity effect (Antov et al., 2012). Chemical coagulants and synthetic inorganic coagulants, such as aluminum salts and polyethylene imine respectively, are widely used coagulants Aluminum residue in drinking water may cause Alzheimer's disease in humans (Bondy 2016). However, the use of inorganic coagulants causes the release of monomers, which are known to be carcinogenic (Zahrim et al., 2011). The maximum acceptable concentration of monomers is 0.1 µg/l according to Commission Directive 98/83/EC, 1998. Chemical coagulants are expensive, produce toxic sludge, and can cause several health problems when used in excess. Nevertheless, alum is extensively used in all water treatment plants in Iraq.

Recently, considerable attention has been paid to developing natural coagulants (Ghernaout 2013). These can be extracted from plant tissue and microorganisms. Natural coagulants should be low cost, safe to human life, eco-friendly, and biodegradable, they should perform well in comparison to chemical coagulants, and they should produce 20-30% less sludge than that of alum remediated coagulant. Several studies have examined the use of natural coagulants, including chitosans (Savi et al., 2016), cacti (Vishali et al., 2014) and chestnuts (S´ciban et al., 2009). In order to extract active coagulants, researchers have used different chemicals at different concentrations. Natural coagulants have shown promise in the purification of water.

Castanea sativa is probably one of the tree species most related with humans. The Humans introduced and managed the chestnut tree as a multipurpose monoculture in many countries. Because only the nuts are used for food processing, other parts like the shells, leaves, and burs represent common waste products of this food industry, even if these residual components contain promising bioactive compounds. (Braga et al., 2014).

Recent studies have shown that by-products and waste from chestnut processing are rich in bioactive compounds with antioxidant, antimicrobial, and anti-inflammatory properties that make them good materials in various fields of application, such as pharmaceuticals, food industry and cosmetics.(Marialuisa et al., 2022)

S´ciban et al. (2009) extracted active coagulant from ground horse chestnut seeds and demonstrated its effectiveness in purifying water. Thus, we were motivated to examine different parts of plants (seeds or leaves) that are abundantly available in Iraq to determine whether effective coagulants could be extracted from them. According to the literature, no previous research has investigated the extraction of natural coagulant from *Castanea Sativa* Tree Leaves (CSTL) in Iraq. Our research aimed to investigate the feasibility of extracting and using natural coagulant from CSTL obtained in Erbil, the capital of the Iraqi Kurdistan Region. We were also interested in investigating the performance of different salts at different concentrations as extracting agents beside distilled water. Jar tests were conducted using synthetic turbid water to adjust turbidity to the required level. Jar tests were also conducted to optimize coagulation conditions, including the dose of coagulant, pH level and initial turbidity. The protein content of CSTL extracts and the impact of natural coagulant on treated water, mainly organic matter content, were also examined.

METHODOLOGY

The feasibility of coagulant extraction from CSTL using distilled water, NaOH, KCl, and NaCl was investigated. Powdered CSTL was also used as a coagulant. Jar tests were conducted to investigate coagulation activity. The effect of the coagulant dosage, pH level and premier turbidity were also examined to optimize the coagulation conditions. The protein content of the bio-coagulants and the possible release of organic matter due to the use of biomaterials were investigated.

The statistical modeling of the practical data obtained was conducted using SigmaPlot 12.5, to affirm the statistical importance related to the data of coagulation. One-way ANOVA was implemented to check for statistical significance at significance level $p < 0.05$.

Analytical methods

The determination of tap water specifications was achieved according to the standard method (APHA, 1998). The turbidity meter (TURB 550 IR) was used to measure turbidity and results were expressed in nephelometric turbidity units (NTU). A pH meter (HQ11d) was used to measure pH levels and electrical conductivity. Protein was thought to be the active coagulant agent; therefore, the Bradford technique was used to estimate the protein content of the extracted coagulants (Bradford, 1976). In the employment of this technique, bovine serum albumin was used as standard. The concentration of organic matter withdraw from the extracted coagulant into the treated water was determined using permanganate index)$KMnO_4$ (method according to the standard method (APHA, 1998). In addition, coagulation results were statistically examined using SigmaPlot 12.5 software.

Natural coagulant extraction

CSTL were collected from Erbil, the capital of Kurdistan Region, Iraqi locate between latitudes 35° 40′ and 36° 30′ N, and longitudes 43° 20′ and 44° 20′ E. The CSTL were dried in an oven at 40 °C before being ground to a fine powder using a coffee mill. The powder was then sieved through 0.35 mm sieve. The powder that was less than 0.35 mm in size was used in this study. To extract the active coagulant component, about 60 g of this powder was dissolved into 1,000 ml of distilled water, KCl, NaCl, and NaOH. The mixture was stirred at room temperature for 30 min using a magnetic stirrer. It was then left to settle for 30 min. The filtrate was purified via a filter of 0.45 µm and clear solution was kept at 4 °C to be used during the experiment. Different concentrations of each salt were used for extraction of the active coagulant. These concentrations were 0.25, 0.5, and 1 M of NaCl and KCl, and 0.025, 0.05, and 0.1 M of NaOH. A flowchart of the extraction process is displayed in Figure 1.

Preparation of synthetic turbid water

Synthetic turbid water was prepared by dispersing kaolin clay (10 g) in tap water (1 liter) and

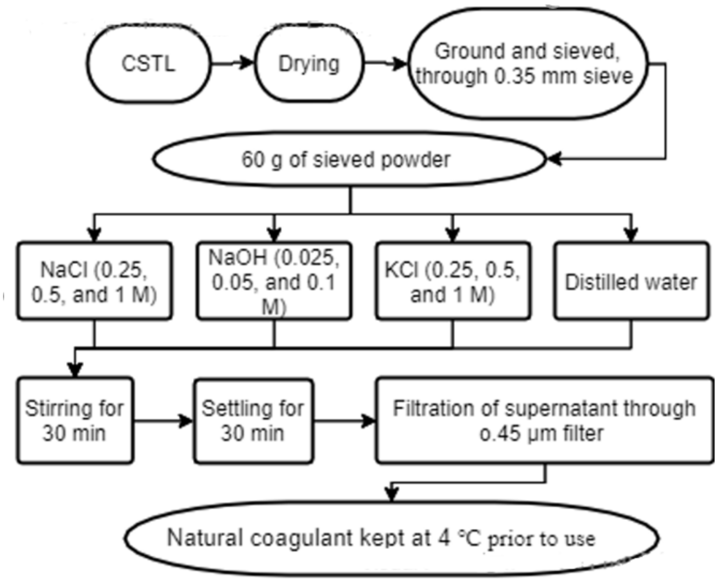

Fig. 1. Flowchart demonstrating the extraction process of CSTL coagulant.

Table 1. Main parameters of tap water used

Parameter	Value
pH	7.6
Turbidity (NTU)	0.43
Electric conductivity (µs/cm)	756
Alkalinity (mg $CaCO_3$/l)	229
Calcium (mg/l)	87
Magnesium (mg/l)	9
Organic matter (mg O_2/l)	1.73

stirring for 2 hr. using the magnetic stirrer. The suspension was removed for 24 h to obtain total hydration. Synthetic turbid water was utilized rather than river water to permit for the alteration of the turbidity level (Choy et al., 2016). The stock solution was kept at 4 °C. The required turbidity level of the specimens was achieved through dilution prior to each jar test. Solution with initial turbidity of 35 NTU was prepared by diluting about 5 ml of stock kaolin solution in 1 liter of tap water. Similarly, water with turbidity level of 17.5 and 70 NTU were prepared by diluting 2.5 and 10 ml of stock solution in 1 liter of tap water (Dhivya et al.,2017). A few drops of 1 M NaOH or 1 M HNO_3 were added to the stock solution to adjust the pH level. The fundamental parameters of tap water used in this study are characterized and listed in **Table 1**.

Coagulation tests

Jar tests were conducted to investigate the performance of the extracted coagulants. 300 ml of synthetic turbid water was added to (6) beakers (600 ml in volume) that were placed in jar test slots and mixed at 200 rpm at room temperature. Different amounts of the extracted coagulants (0.5 to 3 ml/l, with an increment of 0.5 ml/l(were added to the beakers and mixed for 1 min. To initiate the flocculation phase, the speed was decreased to 80 rpm for 30 min. Finally, the suspension was allowed to settle naturally for 1 hr. The supernatant was collected from each beaker and analyzed for residual turbidities and other water quality parameters. A jar test using a blank sample (stoke solution without the addition of the extracted coagulant) was also conducted. The following formula was used to determine coagulation activity. The residual turbidity level in the blank sample was referred to as initial turbidity.

$$Ca(\%) = \frac{It - Rt}{It} * 100 \tag{1}$$

Where; It and Rt are the initial and residual turbidity levels (NTU) respectively and Ca : coagulation activity (%). A jar test using optimum dose and pH was also conducted at different initial turbidities.

RESULTS AND DISCUSSION

Different concentrations of NaCl on coagulant activity were investigated. The coagulant dose, pH and initial turbidity level were also investigated. Coagulation results were statistically examined using SigmaPlot 12.5 software. The results of these investigations are described below.

The impact of salts on the active coagulant

In general, active coagulant can be extracted from seeds or leaves using organic solvents, water, buffer solutions, and different salt solutions (Du et al., 2017). The impact of the extraction agent on coagulants' activity at pH 6 is shown in Figures 2a, b, c, and d. The influence of

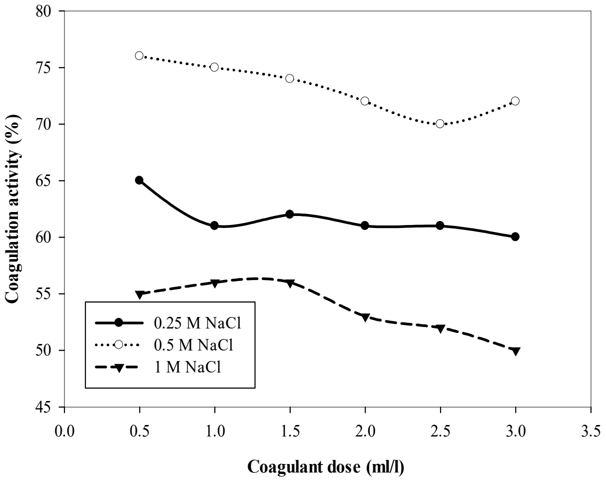

Fig. 2a. Performance of coagulant extracted with different concentrations of NaCl using a range of coagulant doses in the elimination of turbidity at pH 6 and an initial turbidity of 35 NTU.

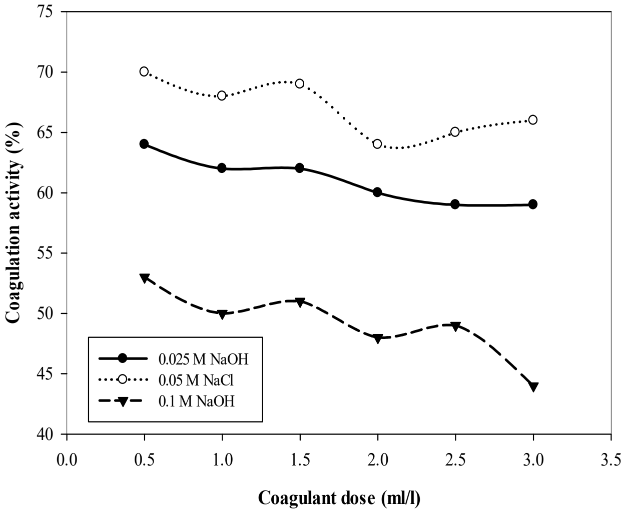

Fig. 2b. Performance of coagulant extracted with different concentrations of NaOH using a range of coagulant doses in the elimination of turbidity at pH 6 and an initial turbidity of 35 NTU.

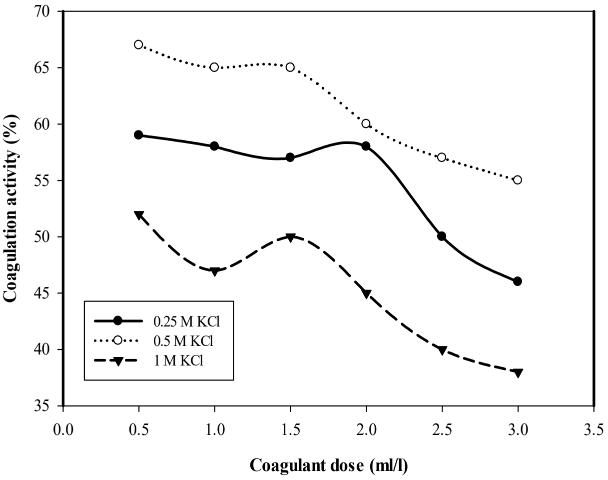

Fig. 2c. Performance of coagulant extracted with different concentrations of KCl using a range of coagulant doses in the elimination of turbidity at pH 6 and an initial turbidity of 35 NTU.

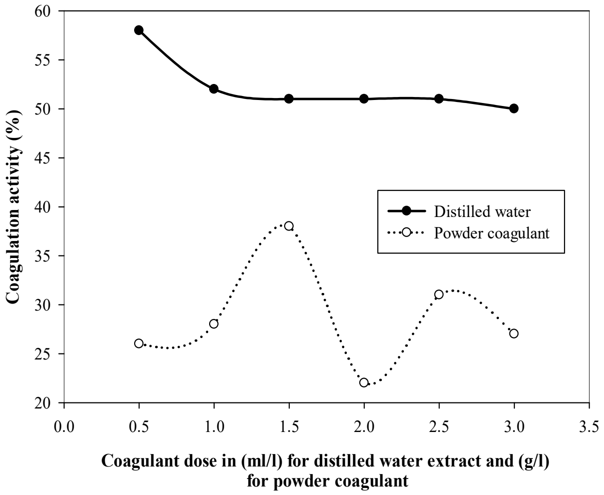

Fig. 2d. Performance of coagulant extracted with distilled water and powdered CSTL using different dosages in the elimination of turbidity at pH 6 and an initial turbidity of 35 NTU.

different doses of three different concentrations of NaCl on coagulant activity was investigated. Results are shown in Figure 2a. The effect of 0.5, 0.25, and 1 M of NaCl on coagulant activity was 76%, 65% and 55% respectively, using a dose of 0.5 ml/l (Figure 2a).

Coagulation activity was significantly higher using 0.5 M NaCl than that using 0.25 and 1 M NaCl (Figure 2a). The efficiency of coagulant extracted with 0.25 M NaCl was significantly better than that of coagulant extracted with 1 M NaCl as the coagulant extracting agent (Figure 2a). Increasing NaCl concentration from 0.25 to 0.5 M resulted in significant improvement in the performance of the extracted coagulant. The influence of NaCl can be related to two phenomena known as the salting-in effect and the salt effect (Huang et al., 2017).

Increasing NaCl concentration from 0.25 to 0.5 M causes more active coagulant extraction from CSTL, which dissolve in NaCl. Thus, increasing the concentration of salt (NaCl) to 0.5 M boosts protein solubility in the aqueous solution, which is known as the salting-in effect (Dhivya et al., 2017). The salt effect relates to increasing ionic strength resulting in an increase in particle aggregation. This usually occurs due to double layer compression. Increasing NaCl concentration over 0.5 M significantly reduces the effectiveness of the extracted coagulant due to the salting-out effect and the hydration effect (Folens et al., 2017). The salting-out effect is the reduction of protein solubility when salt concentration (NaCl) rises above 0.5 M (Du et al., 2017). The influence of different concentrations of NaOH on the extracted coagulant can be investigated by observing its effectiveness at water purification (Figure 2b). Increasing NaOH concertation up to 0.05 M significantly improved the ability of the extracted coagulant to eliminate turbidity (Figure 2b).

Any further increase in the concentration of NaOH had an adverse effect on the performance of the extracted coagulant. This is caused by the moderated solubility of the protein due to protein denaturation (Foroughi et al., 2018). The influence of different concentrations of KCl on the performance of the extracted coagulant followed a similar pattern to that of NaOH and NaCl at different concentrations (Figure 2c).

The performance of coagulant extracted with distilled water was significantly higher than the performance of powdered CSTL (Figure 2d). The highest level of turbidity removal using distilled water as an extracting agent was about 58%, at a coagulant dosage of 0.5 ml/l (Figure 2d), whereas the highest turbidity removal using the powdered CSTL was 38% at 1.5 g/l of powdered coagulant. In order, the best coagulants of all the extracting agents were 0.5 M NaCl, 0.05 M NaOH, 0.5 M KCl, distilled water, and powdered coagulant.

Impact of coagulant dose

The coagulant dose is a crucial factor in the cogulation process (Teh et al., 2014). This is due to the strong correlation between bivalent cations and the dose of coagulant. In general, increasing the coagulant dose had a negative impact on coagulation activity (Figures 2a, b, c, and d). Coagulation activity reduced slightly as the coagulant dose increased. For example, increasing the coagulant dose from 0.5 to 3 ml/l significantly reduces coagulant activity from 67% to 55% when employing 0.5 M KCl as an extracting agent (Figure 2c). The excess of coagulant amount over the required dosage, i.e. (over-dose) which may have resulted from the steric stabilization, perhaps lead to degradation due to the relation between the degradation and steric stabilization. (Kukić et al., 2015).

No statistical significance was observed in the performance of coagulants using dosages of 0.5 and 1 ml/l (Figure 2a, b, c and d). Generally, a coagulant dosage of 0.5 ml/l was most effect for distilled water and salts extract, while a dose of 1.5 g/l was most effect for powdered CSTL (Figure 2a, b, c and d). Using a small amount of bio-coagulant is highly beneficial as biomaterials may contribute to organic matter content in treated water, which leads to microbial growth. Moreover, there are economic benefits to using lower quantities of coagulant.

The impact of pH

Several parameters, including the initial turbidity level, jar test conditions, the coagulant dose, initial turbidity, pH, and the nature of the coagulant, can affected by the coagulation process. The effect of pH level on coagulant performance was investigated within the range of 5.5 to 8. This was to maintain a minor difference from the upper admissible pH level of 6.5 to 8.5, according to Iraqi standards. The impact of pH on the elimination of turbidity using all concentrations of NaCl, KCl, and NaOH are presented in Figures 3, 4, and 5 respectively.

Figure 6 shows the correlation between pH level and coagulation activity using the powder coagulant and distilled water as extracting agents. The pH level can affect the charge of protein molecules; therefore, it affects coagulation process (Liu et al., 2017). Increasing the pH level from 5.5 to 8 resulted in a significant improvement in turbidity removal efficiency (Figures 3, 4, 5, and 6). This can be related to the influence of pH on protein. At high pH levels, protein is expected to have anionic in CSTL extracts (Witek-Krowiak et al. 2014). Consequently, a net-like structure is created between negatively charged proteins that attracts bivalent. This eventually causes the kaolin suspension to become trapped in the net and settled out of the solution. These results are in accordance with the findings of Antov et al. (2012) that high

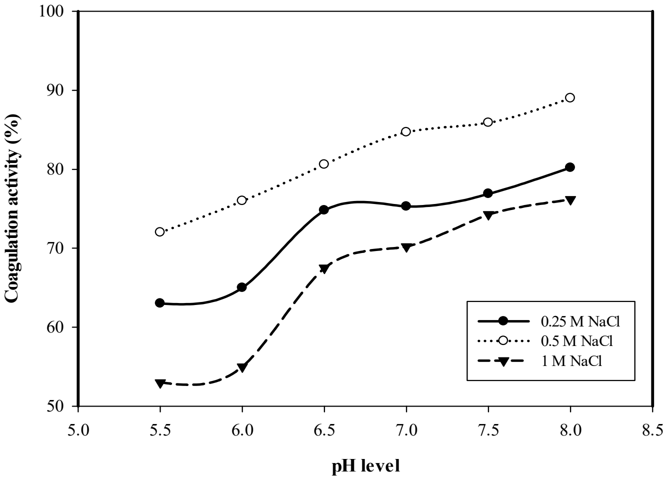

Fig. 3. The impact of pH on the performance of coagulant extracted with different concentrations of NaCl, using a dose of 0.5 ml/l, with initial turbidity of 35 NTU.

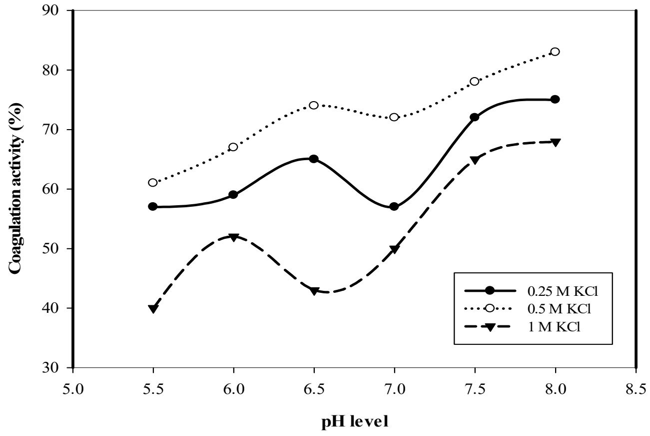

Fig. 4. The impact of pH on the performance of coagulant extracted with different concentrations of KCl, using a dose of 0.5 ml/l, with initial turbidity of 35 NTU.

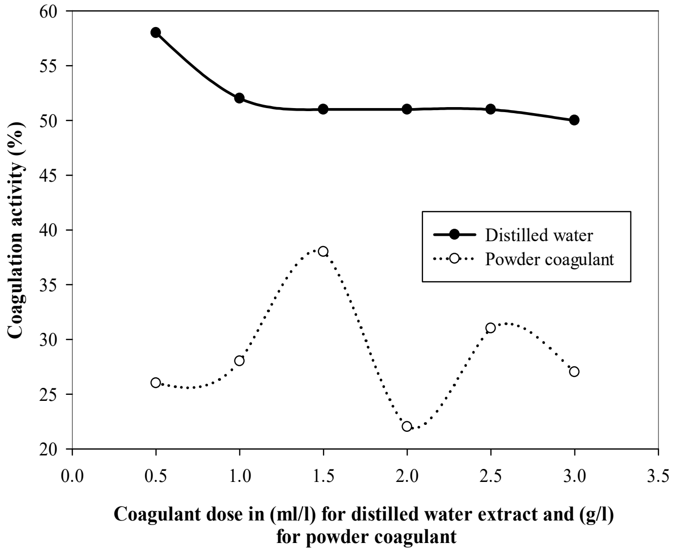

Fig. 5. The impact of pH on the performance of coagulant extracted with different concentrations of NaOH, using a dose of 0.5 ml/l, with initial turbidity of 35 NTU.

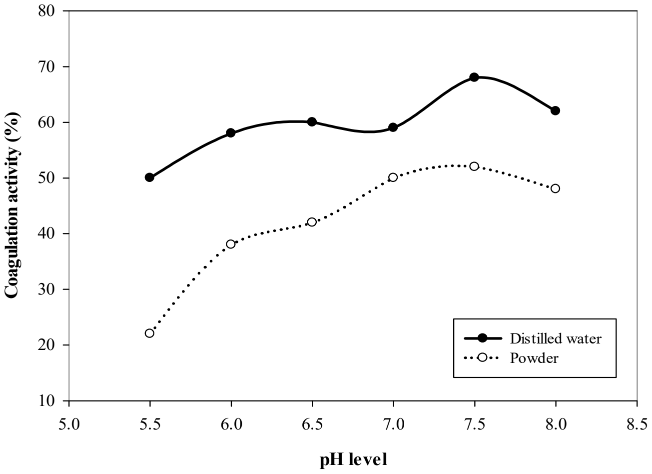

Fig. 6. The impact of pH on the performance of coagulant extracted with distilled water, using a dose of 0.5 ml/l, and the powder coagulant, using a dose of 1.5 gm/l, with initial turbidity of 35 NTU.

pH levels are best for coagulation. Powdered CSTL and coagulants extracted with distilled water performed best at pH level of 7.5 (Figure 6). Figure 7 compares the performance of coagulants extracted using NaCl, NaOH, KCl at their most effective concentrations, as well as distilled water and the powdered CSTL. The coagulant extracted with NaCl and NaOH followed a similar trend (Figure 7). The order of coagulants based on their performance is as follows: NaCl, NaOH, KCl, distilled water, and powder coagulant (Oladoja, 2015). This may be related to the ability of NaCl to extract more protein due to salt in effect that increases protein solubility, which believed to be the active coagulant (Subramonian et al., 2014).

The impact of the initial turbidity level

The value of initial turbidity and its effect was examined for coagulants extracted with NaCl, KCl, and NaOH due to their good activity in comparison to distilled water extract and powdered CSTL. The impact of initial turbidity on NaCl, KCl, and NaOH at pH 8 using dose of 0.5 ml/l is shown in Figure 8, 9, and 10. Higher coagulation activity were achieved at initial turbidity level

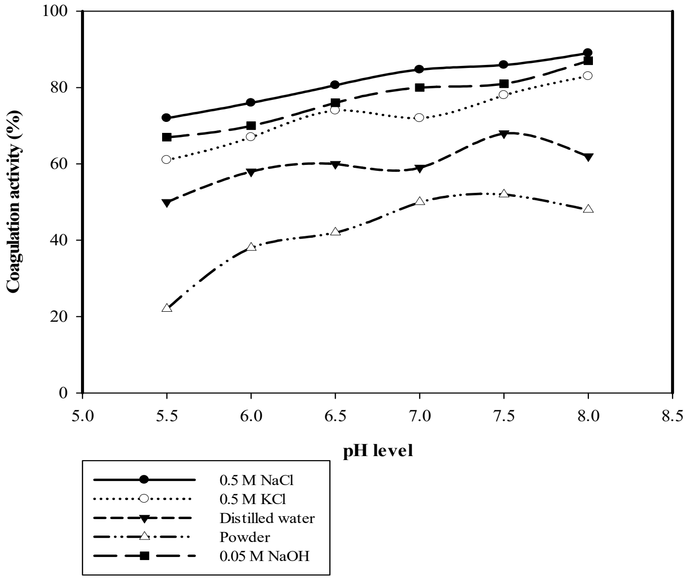

Fig. 7. Comparison between performance of coagulants extracted with 0.5 M NaCl, 0.5 M KCl, 0.05 M NaOH, distilled water, and powdered CSTL with a pH ranged of 5.5 to 8, initial turbidity of 35 NTU, and a coagulant dose of 0.5 ml/l, for powdered CSTL only dose of 1.5 g/l.

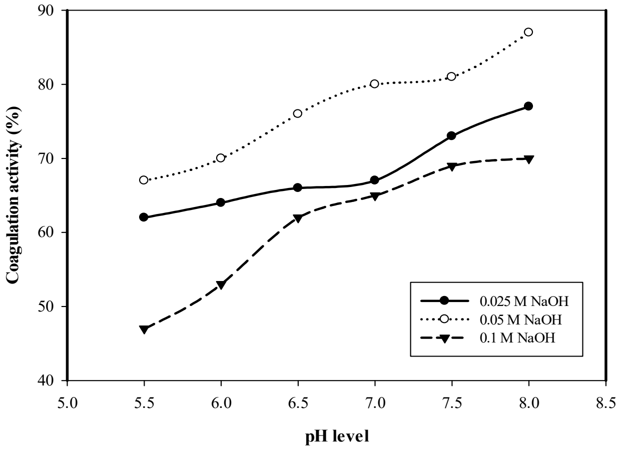

Fig. 8. The impact of initial turbidity on the performance of coagulant extracted with NaCl using a dose of 0.5 ml/l at pH 8.

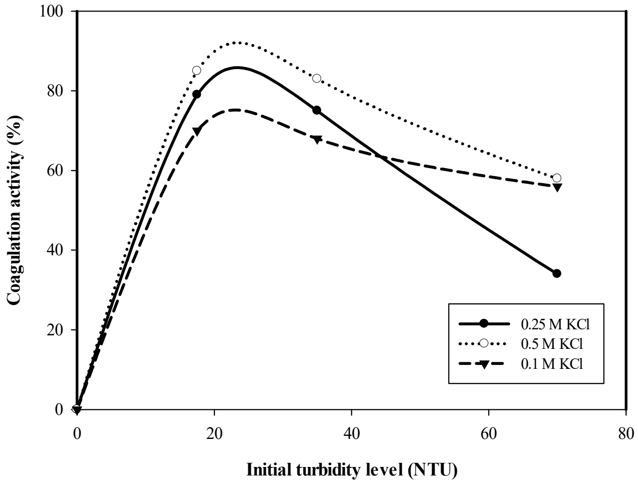

Fig. 9. The impact of initial turbidity on the performance of coagulant extracted with KCl using a dose of 0.5 ml/l at pH 8.

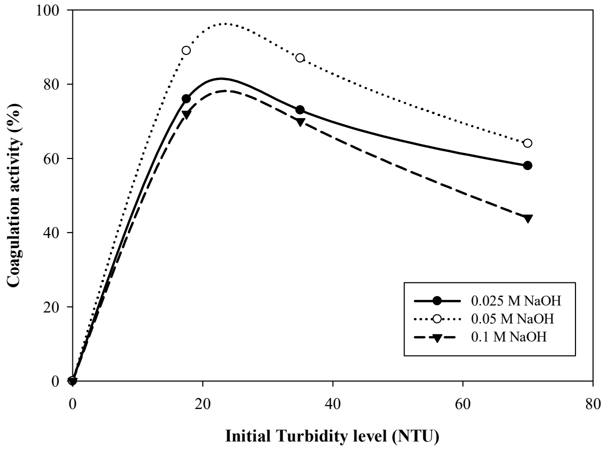

Fig. 10. The impact of initial turbidity on the performance of coagulant extracted with NaOH using a dose of 0.5 ml/l at pH 8.

of 17.5, while lowest activity was observed at turbidity level of 70 NTU (Figures 8, 9, and 10). This indicates that the studied parameters are appropriate for treating water with initial turbidity equal or less than 35 NTU.

Protein content

It has been suggested that protein plays an active role in the process of coagulation when using natural coagulants (Choy et al., 2016). The concentrations of protein within the extracted coagulant were determined at the most effective concentration of salts, that is, 0.5 M NaCl, 0.5 M KCl, and 0.05 M NaOH (Figures 2a, b, and c). Protein content in distilled water extract and powdered CSTL was not examined due to its moderate performance in the elimination of turbidity (Figure 2d). The amounts of protein extracted are displayed in Table 2. These were 0.322, 0.283, and 0.274 mg/ml using 0.05 M NaCl, 0.5 M NaOH, and 0.5 M KCl, respectively (Table 2).

The amount of protein content of the 0.5 M NaCl was greater than that of the 0.05 M NaOH and 0.5 M KCl (Table 2). This shows that the coagulation activity of extracted coagulants were proportional to protein content (Shamsnejati et al., 2015). However, protein content of NaOH is

Table 2. Amount of protein (mg/ml) in coagulants extracted with different salts

Extracting agent	Protein content (mg/ml)
0.5 M NaCl	0.322
0.05 M NaOH	0.283
0.5 M KCl	0.274

Table 3. The amount of organic matter in water treated with CSTL extracts, at a coagulant dose of 0.5 ml/l, initial turbidity 35 NTU, and pH 8.

Coagulant	Organic matter (mg O_2/l)	Turbidity (NTU)	Electric conductivity (µs/cm)
0.05 M NaOH	2.27	4.5	789
0.5 M NaCl	2.81	3.8	793
0.5 M KCl	2.21	5.9	787

only slightly greater than that of KCl extrat. This can be attributed to the fact that the coagulant extract constitutes an aqueous solution that contains not only protein, but also different types of polyelectrolytes (Wu et al., 2016). Polyelectrolytes can act as a natural coagulant.

The impact of natural coagulant on organic matter content

The main characteristics of treated water are displayed in Table 3. If natural organic materials are introduced into water, they can negatively affect water quality, primarily by increasing organic matter content. High concentrations of organic matter can cause microbiological issues while water is being stored, as well as undesirable taste, color, and odor (Shokoohi et al., 2015). Organic matter can also lead to an increase in the amount of chlorine that must be used in water treatment plants. Therefore, the presence of organic matter was examined after coagulation process. Organic matter content, and electrical conductivity were measured for water coagulated at pH 8, an initial turbidity of 35 NTU, and a coagulant dose of 0.5 ml/l for coagulants extracted with 0.5 M NaCl, 0.5 M KCl, and 0.05 M NaOH. The concentration of organic matter in the tap water sample was about 1.73 mg O_2/l (Table 1).

Organic matter content slightly increased to 2.81 mg O_2/l when using 0.5 M NaCl as the extracting agent (Table 3). Coagulants extracted with 0.05 M NaOH and 0.5 M KCl respectively increased organic matter to 2.27 and 2.21 mg O_2/l (Table 3). The issue of increasing organic matter in the treated water should be carefully attained, due to the relation with the protein content of the extracted coagulant (see Table 2). The increase in organic matter could be related to the solubility of organic matter that did not settle out of the solution during coagulation (Zia et al., 2015). According to Iraqi standards for drinking water, the maximum admissible level of turbidity and electrical conductivity are 5 NTU and 1,563 µs/cm respectively. Both the 0.5 M NaCl and 0.05 M NaOH extracted coagulants successfully reduced turbidity to below 5 NTU (Table 3). For the 0.5 M KCl extracted coagulant, the turbidity level was reduced to about 5.9 NTU, which is slightly higher than the upper admissible level, according to Iraqi standards (Table 3). Hence, for water with initial turbidity of 17.5, turbidity level was reduced to below admissible levels using 0.5 M NaCl, 0.05 M NaOH, and 0.5 M KCl extracts.

CONCLUSIONS

The feasibility of coagulant extracted from CSTL in the treatment of turbid water was investigated using several salts. Jar test was performed to detect the performance of the new extracted coagulants. Powdered CSTL was also used as a natural coagulant. The extracting agents that performed best were, in order, 0.5 M NaCl, 0.05 M NaOH, 0.5 M KCl, distilled water, and the powder coagulants. The greatest amount of protein was observed in the NaCl extract. Generally, increasing the dose of coagulant moderately reduces the effectiveness of the coagulants. The optimum dose was 0.5 ml/l. In addition, at high pH levels there was a significant improvement in turbidity elimination. The optimum pH in this study was 8 for NaCl, NaOH, and KCl extracts. Bio-coagulant lead to unpretentious increasing in organic matter in treated water. Further research is required to examine the feasibility of CSTL in raw wastewater coagulation.

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The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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