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Turbidity removal from aqueous environments by *Pistacia atlantica* (Baneh) seed extract as a natural organic coagulant aid

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ABSTRACT

Coagulation/flocculation, as an important step in water treatment processes, is applied for turbidity removal of raw water coming from suspended particles and colloidal material. In this study, the ability of *Pistacia atlantica* seed extract, as a natural organic coagulant aid along with ferric chloride for turbidity removal from synthetic solutions, was investigated. Coagulation ability of this coagulant was assessed by the use of standard jar test measurement. The influence of several operating parameters such as, pH (2–12), coagulant dose (2–35 mg/L), natural coagulant aid dose (0.1–5.0 ml/L), and initial turbidity (20–200 NTU), were investigated. The optimum pH for removal of turbidity by coagulation process using *P. atlantica* seed extract was found 9–10 with coagulant dose 1 ml/L (in combination with $FeCl_3$ at dose 2 mg/L). The results obtained with synthetic solutions revealed that the most effective removal efficiency of turbidity (97.43%) was achieved with natural coagulant dose 0.4 ml/L and $FeCl_3$ 10 mg/L at pH 9. In addition, the highest removal efficiency was obtained at higher initial turbidities. According to the results of this study and with respect to the abundance of this tree in various regions of Iran, especially in mountain areas, its use as a natural coagulant for turbidity removal from aqueous environments is possible and viable specifically for rural areas and small communities.

Keywords: Natural coagulant; Turbidity removal; *Pistacia atlantica*; Coagulation

1. Introduction

In water and wastewater treatment, coagulation and flocculation process are used for removal of turbidity, suspended solids, natural organic matters, heavy metals, and some anions [1,2]. Many coagulants and flocculants are extensively used in conventional water treatment processes. These materials can be classified into inorganic coagulants (e.g. aluminum and ferric salts) and synthetic organic polymers (e.g. polyacryl amide derivatives and polyethylene amine).

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Alum, ferric chloride, and sulfate were commonly used [3]. Unfortunately, in many developing countries, the cost of importing alum, poly aluminum chloride (PACl), ferric chloride, and other required chemicals for conventional water treatment may be too high and at times, prohibitive. There is also the possibility that a secondary pollution problem will arise due to excessive chemical use. Furthermore, the application of these chemical coagulants in water treatment process has some limitations such as ineffectiveness in low-temperature water, relatively high procurement costs, detrimental impacts on human health, production of large sludge volumes, and the fact that they considerably affect the pH of treated water. There is also strong evidence linking aluminum-based coagulants to the development of Alzheimer in human beings. Alternative coagulants, preferably natural and locally grown, might be economically viable [4].

Thus, in water treatment process, the use of natural coagulants could be an alternative with many advantages over chemical agents, particularly the biodegradability, low toxicity, and fewer risks to health and environment and also, low residual sludge production [5]. Indeed, natural coagulants of vegetable and mineral origin were in use in water treatment before the advent of chemical salts as coagulant, but they have not been able to compete effectively due to the lack of a scientific understanding of their effectiveness and mechanism of action. Thus far, the use of natural coagulants has been discouraged without any scientific assessment. They have progressively succumbed under modernization and survived only in remote areas of some developing countries [4–6]. Recently, however, there has been a resurgence of attention in natural coagulants for water treatment in developing countries, mainly due to the mentioned reasons.

Biopolymers may be of great interest because they are natural low-cost products, characterized by their environmental-friendly behavior and typically have large number of surface charges that increase the effectiveness of the coagulation process. These advantages improve, especially when the plant from which the coagulant is extracted is indigenous to local community. In recent numerous studies, the variety of plant materials has been reported as a source of natural coagulants [6–9].

Chestnut and acorn seed extracts [9], *Moringa peregrine* seed [10], *Pistacia atlantica* [11], Cactus [12], *Moringa oleifera* seed [13], tannin [14], *Acacia mearnsii* [15], and *Cassia angustifolia* seed [16] are some of the natural coagulants used for turbidity, suspended solids, color, and other pollutants removal from aqueous solutions.

Iran is the largest producer of pistachio (*Pistacia* spp.) in the world, with over 44% of global production [17]. Most of the production is from orchards that account for 53% of world planted area, nevertheless there are a few places, such as in the Zagros Mountains, where wild pistachio (*P. atlantica Desf.*) persists in natural and expansively managed (i.e. semi-natural) stands. Wild pistachio, known as Baneh in Iran, is the most economically important species for rural communities in natural forest areas. Wild pistachio trees cover an area more than 1,200,000 ha mainly in the western, central, and eastern parts of Iran. Cultivation of pistachio for multiple uses is believed to have been practiced in Iran for perhaps 3,000–4,000 years [17,18]. The fruit of wild pistachio is an important source of food (after grinding and mixing with other ingredients), although the fruit is smaller and not as commercially valuable as those produced in orchards (primarily from cultivars of *Pistacia vera L.*) and also, the average production of pistachio seeds for each tree is about 80–120 kg per year [18,19].

The objective of the current study is to experimentally evaluate the efficiency of *P. atlantica* seed extract as a natural coagulant aid in water turbidity removal. Coagulation efficiencies of prepared coagulant were measured at different pH values and dosages.

2. Materials and methods

2.1. *Preparation of P. atlantica* seed extract

Dry *P. atlantica* seeds were collected in the vicinity of Kashmar city (35˚12´N, 58˚30´E), Khorasan Razavi province, Iran. The preparation and extraction process was carried out in the following way: The seeds coats and wings were removed, and seeds was ground to a fine powder by using a laboratory mill and sieved through 0.4 mm sieve. A 1-M NaCl solution was prepared and 5 g of powder were put into 100 ml of it. The NaCl solution with powder was stirred for 30 min at room temperature (around 20 ± 2 ℃) to extract the coagulation active components. No pH modification was needed as natural pH 7 was achieved. Then, the extract was filtered twice: once through commercial filter paper on Buchner funnel and once through a fine filtering Millipore system (0.45 μm glass fiber). The result is a clear, milk-like liquid. To prevent any aging effect, such as changes in pH, viscosity, and coagulation activity due to microbial decomposition of organic compounds during storage, fresh coagulant agent was prepared and used immediately for each sequence of the experiment.

2.2. Solutions preparation (preparation of turbid water)

All the chemicals used were of analytical reagent grade and were purchased from Merck Company (Germany). Turbid water for coagulation tests was prepared by adding 2 g kaolin to 1 L distilled water. The suspension was stirred for 2 h to achieve a uniform dispersion of kaolin particles, and then it was allowed to remain for 24 h for completing the hydration of the particles. This suspension was used as the stock suspension. Working solutions of the desired concentrations of turbid water (10–200 NTU) were obtained by successive dilutions just before the coagulation test. The initial pH of the synthetic water was adjusted to desired values (2–12) with 1 mol/L NaOH or HCl right before experiments. The pH of solution was measured using a pH meter (model E520, Metrohm Herisau, Switzerland). The ranges of experimental parameters are presented in Table 1. All the solutions were prepared in double distilled water.

2.3. Coagulation experiments

The coagulation–flocculation experiments were conducted in a conventional jar test apparatus (VELP-Scientifica, Model: JLT6, Italy) with impellers equipped with 2.5 × 7.5 cm rectangular blades and six 1,000 ml beakers with respect to the effect of pH, dosage of the main coagulant, and aid coagulant and also, the initial turbidity of samples. The operating parameters used were based on the reviewed literature. Six 1 L beakers were filled with 1,000 ml of predetermined turbidity concentration of solutions and placed in the slots of a jar tester which was equipped with an illuminator. Various dosages of ferric chloride ($FeCl_3$) and *P. atlantica* seed extract were added to each beaker and agitated for 4 min at 100 rpm for rapid mixing. The mixing speed was reduced to 30 rpm for another 15 min. All suspensions were then left for sedimentation. After 30 min of sedimentation, the clarified samples were collected from the top of the beaker and filtered to remove any remaining sediment. The turbidity of each clarified sample was then measured using a turbidimeter (Hach Turbidimeter Model 2100 N). In addition, concentration of suspended solids was determined according to the standard methods for the examination of water and wastewater [20]. To survey the effect of pH parameter on turbidity removal, pH of solution was varied from 2–12, while the dosages of *P. atlantica* seed extract and $FeCl_3$ were studied from 0.1 to 5 ml/L and 2 to 35 mg/L, respectively. The initial turbidity was investigated from 10 to 200 NTU. All experiments were performed at the room temperature (21 ± 2 ˚C) and carried out in duplicate for consistency, and the average results are presented herein.

In the end, a series of jar tests were performed using the 1 L of natural water (real sample) with optimum conditions attained. The characterization of the natural water used for these experiments is listed in Table 2.

3. Results and discussion

3.1. Effect of initial pH

Since pH is a very important parameter regarding both coagulation process and charges on protein molecules, the influence of pH on coagulation activity was investigated firstly [21,22]. Fig. 1 shows the effects of initial pH values within the range of 2–12 on the efficiency of turbidity removal by *P. atlantica* seed extract with initial dosage of 1 ml/L and $FeCl_3$ dosage of 2 mg/L at two initial turbidity concentrations of 20 and 50 NTU. Results showed that, the most appropriate pH for both turbidity concentrations of 20 and 50 NTU was found to be pH 10 (equal to 69.5% of removal efficiency) and 9 (equal to 78.7% of removal efficiency), while in pH range from 2 to 4, the minimum removal efficiency was attained (Fig. 1). On the other hand, turbidity removal efficiency increased with increasing pH. In fact, according to the results of this study (Fig. 1), the optimum pH for maximum removal of turbidity was found 9 (78.7% removal of turbidity), and removal efficiency at pH 7–8 was about 74.5–75.7%. Consequently, with respect to pH of natural waters (in the rages 6.5–8.5), adjustment of pH is very simple and it can be performed by adding lime or soda ash at very low doses.

Table 1
The ranges of experimental parameters

pH	Coagulant dosage (mg/L)	Natural coagulant dose (ml/L)	Rapid mixing, slow mixing, and sedimentation time	Initial turbidity, NTU
2–12	2, 4, 6, 8, 10	0.08, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 2.0, 3.0, 4.0, 5.0	4 min (100 rpm), 15 min (30 rpm), and 30 min	10, 20, 50, 80, 100, 200

Table 2
The raw water quality characteristics used for the some experiments

Parameter	Concentration
pH	7.71
Conductivity, µS/cm	683
Total dissolved solids, mg/L	452.6
Turbidity, NTU	14.6
Iron, mg/L	0.04
Manganese, mg/L	0.012
Sulfate, mg/L	41.2
Chloride, mg/L	47.9
Sodium, mg/L	28.1
Nitrate, mg/L	6.4
Nitrite, mg/L	0.03

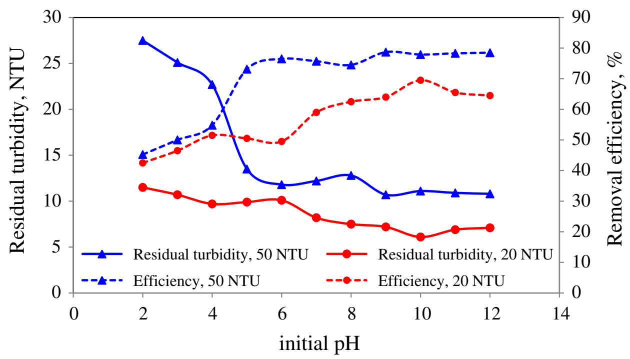

Fig. 1. Effect of pH on turbidity removal from synthetic solutions (*P. atlantica* seed extract dose = 1 ml/L and $FeCl_3$ dosage = 2 mg/L).

Turbidity removal in the alkaline pH range is presumably due to the adsorption onto hydroxide flocs and also as a result of precipitation, co-precipitation, and adsorption mechanisms. These findings are adequately in accordance with observations that higher pH values are optimal for other natural coagulants e.g. from *M. oleifera* [23], from *M. peregrine* [24], from *P. juliflora* and *C. latifaria* [7], from *C. angustifolia* [16], or common bean [25]. In addition, the results obtained by Zhang et al. on turbidity removal by cactus as coagulant showed that the optimum pH was about 10, and the worst effect appeared for pH about 6 [25]. It can be concluded that natural coagulants are most effective at basic waters as evident by the optimum pH values from 7 to 10 [26].

In addition, the results showed that the extract of *P. atlantica* seed has a minimal effect on the pH of the samples. This result is in agreement with those results obtained by Ndabigengesere et al. on removal of turbidity by *M. Oleifera* seed extract [27], and also by Bazrafshan et al. on arsenic removal by *M. peregrine* seed extract [10]. Moreover, they reported that the efficiency of the *M. Oleifera* extract as a coagulant is not affected by pH.

3.2. Effect of combined $FeCl_3$ and natural coagulant concentration on turbidity removal

Coagulant dosage is one of the major parameters that have been considered to determine the optimum condition for the performance of coagulants in coagulation and flocculation processes. Basically, insufficient dosages or overdosing may result in a poor performance in flocculation. Consequently, it is essential to determine the optimum dosage of coagulant in order to minimize the dosing cost and sludge formation and also to obtain the optimum performance in treatment. Therefore, the effect of coagulant dose of *P. atlantica* seed extract on turbidity removal at various doses of $FeCl_3$ (2–10 mg/L) was depicted in Fig. 2, showing that coagulation efficiency of turbidity changes with an increase in dosage of *P. atlantica* seed extract from 0.08 to 5 ml/L. The optimum dosage of *P. atlantica* seed extract at different doses of $FeCl_3$ was different. On the other hand, the highest turbidity removal was found to be 95.28, 95.9, 96.43, 97.08, and 97.43% at natural coagulant dosages equal to 0.9, 0.4, 0.4, 0.2, and 0.4 ml/L, at $FeCl_3$ doses 2, 4, 6, 8, and 10 mg/L, respectively. More increase in the dosage of natural coagulant did not improve the removal of turbidity. In addition, as shown in Fig. 2, turbidity removal efficiency increased with an increase in the dosage of $FeCl_3$, so the maximum efficiency was achieved at coagulant dose 10 mg/L. Therefore, at present study, coagulant dosage, 0.4 ml/L, was selected as the

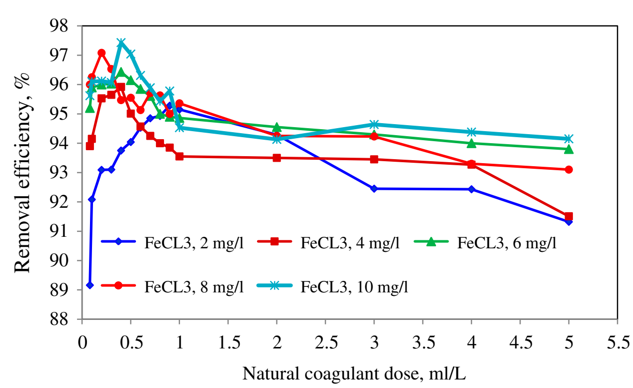

Fig. 2. Effect of coagulant dose (*P. atlantica* seed extract) on turbidity removal from synthetic solutions (at optimum pH 9 and initial turbidity ~100 NTU).

optimum dose of coagulant. Obtained results for optimum pH values of coagulant are in good agreement with the values given in [16,28]. On the other hand, over the optimal dose, the excess of the flocculating agent would disturb sedimentation and cause the resuspension of the aggregated particles [29].

Generally, it can be concluded that lower doses of investigated natural coagulants were better than higher ones. This is very important not only for process economy but also for lower organic matter load in processed water because it is known that high organic load might cause microbial growth [9]. Nevertheless, obtained results are valid only in applied conditions. Finally, it can be concluded that removal of turbidity is achieved by water-soluble cationic proteins in the *P. atlantica* seed extract, which enhance both coagulation and flocculation.

In addition, the results, as shown in Fig. 3, indicate that, as the coagulant dose increased from 0.1 to 1 ml/L, the turbidity of the solution reduced significantly. A further increase in coagulant dose decreases the turbidity removal efficiency. The best efficiency for initial turbidity 10, 20, 50, 80, 100, and 200 NTU was obtained at coagulant dose 2 (68.5%), 3 (69.8%), 1 (70.4%), 2 (72.5%), 1 (75.2%), and 1 ml/L (78.4%), respectively. The increasing and decreasing trend in the turbidity removal can be explained by the fact that, for up to 1–2 ml/L, the increase of coagulant dose enhances the aggregation and consequently, the particle settling. However, over the optimal dose, the excess quantity of the natural coagulant might disturb sedimentation and cause the redispersion of the aggregated particles [29]. Similar findings were reported by other researchers [7,24,29].

3.3. Effect of initial turbidity on turbidity removal by *P. atlantica* seed extract as a natural coagulant

Fig. 3 depicts the effect of initial turbidity values within the range of 10–200 NTU on the efficiency of *P. atlantica* seed extract at dosages of 0.1–5.0 ml/L (without adding $FeCl_3$). As it can be seen from Fig. 3, with increase of initial turbidity, its removal efficiency increased, too. For example, at natural coagulant dose 1 ml/L, turbidity removal efficiency for initial turbidity 10 NTU was 66.9% and with increasing the initial turbidity to 100 and 200 NTU, removal efficiency increased to 70.4 and 77.3%, respectively. This was in agreement with findings reported by other researchers [12,30]. They documented the increase in turbidity removal with increasing the initial turbidity of a sample.

3.4. Experiments on raw water

Some experiments were performed to study the efficiency of *P. atlantica* seed extract as natural coagulant at different dosages for turbidity removal from raw water samples (at the point of entry to water treatment plant) at optimum pH 9 and initial turbidity 14.6 NTU. The characteristics of raw water used for these experiments are presented in Table 2. As it can be seen in Fig. 4, the removal efficiency of turbidity increased with increase in natural coagulant concentration, so maximum efficiency was achieved at natural coagulant dose 3 ml/L (71.4%). Nevertheless, more increase of natural coagulant decreased turbidity removal efficiency. In addition, as shown in Fig. 4, dual application of *P. atlantica* seed extract and $FeCl_3$ resulted in better removal of turbidity (78.6%).

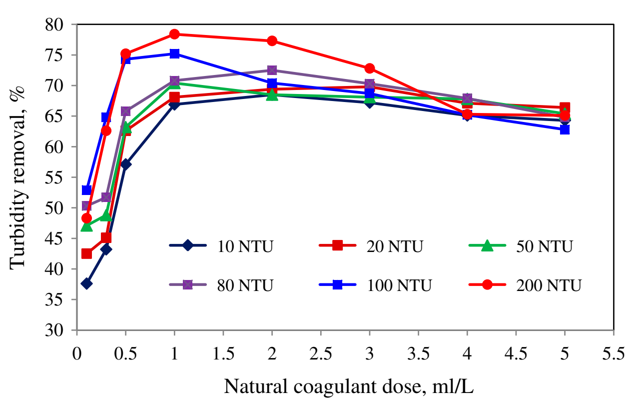

Fig. 3. Effect of altering the dosage of the *P. atlantica* seed extract with initial turbidities of 10, 20, 50, 80, 100, and 200 NTU (initial pH 9).

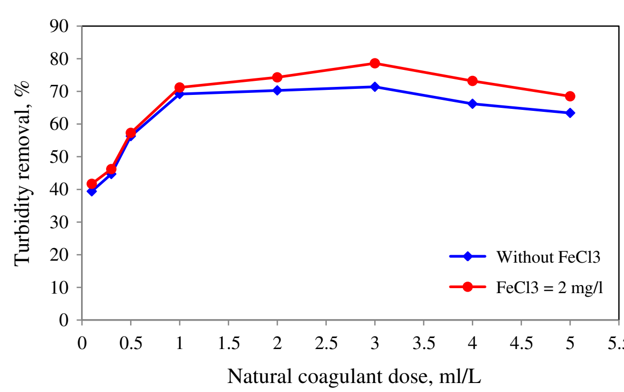

Fig. 4. Effect of altering the dosage of the *P. atlantica* seed extract on turbidity removal from raw water (initial pH 9 and turbidity = 14.6 NTU).

Muyibi and Okuofu [30] used the extract of *M. oleifera* seeds to treat waters obtained from three surface water sources in Nigeria; the result was that turbidity removal efficiency could reach 50%. The optimum dosage of the seeds of *M. Oleifera* was comparable to that of alum. As it can be seen from Fig. 4, turbidity removal efficiency with *P. atlantica* seed extract is comparable to $FeCl_3$, especially at low doses of natural coagulant. Similar findings were observed for removal of suspended solids from real samples (data not presented). Furthermore, final pH of treated water with *P. atlantica* seed extract as natural coagulant aid and $FeCl_3$ was about 8.3–8.5; hence, according to current criteria for drinking water, pH adjustment is not necessary. Consequently, it can be concluded that *P. atlantica* seed extract can be used for turbidity removal from natural waters.

4. Conclusion

In the present study, the efficiency of natural coagulant obtained from *P. atlantica* seeds in water turbidity removal was experimentally evaluated. Coagulation efficiency of prepared coagulant extract was measured for different pH values and dosages. The results obtained with synthetic solutions revealed that the most effective removal efficiency of turbidity (97.43%) was achieved with natural coagulant dose 0.4 ml/L and $FeCl_3$ 10 mg/L at pH 9. In addition, the highest removal efficiency was obtained at higher initial turbidities. Finally, according to the achieved results, it can be concluded that *P. atlantica* seed extract is an efficient coagulant for turbidity removal from aqueous environments and use of *P. atlantica* seed extract as a coagulant aid may play a key role in reducing the amount of $FeCl_3$ as the main coagulant and consequently decreasing the generated sludge. Furthermore, due to abundance of this tree in various regions of Iran, especially in mountain areas, its use as a natural coagulant is possible and viable, especially for rural areas.

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