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Assessment of groundnut (*Arachis hypogaea*) as a natural coagulant for water treatment

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ABSTRACT

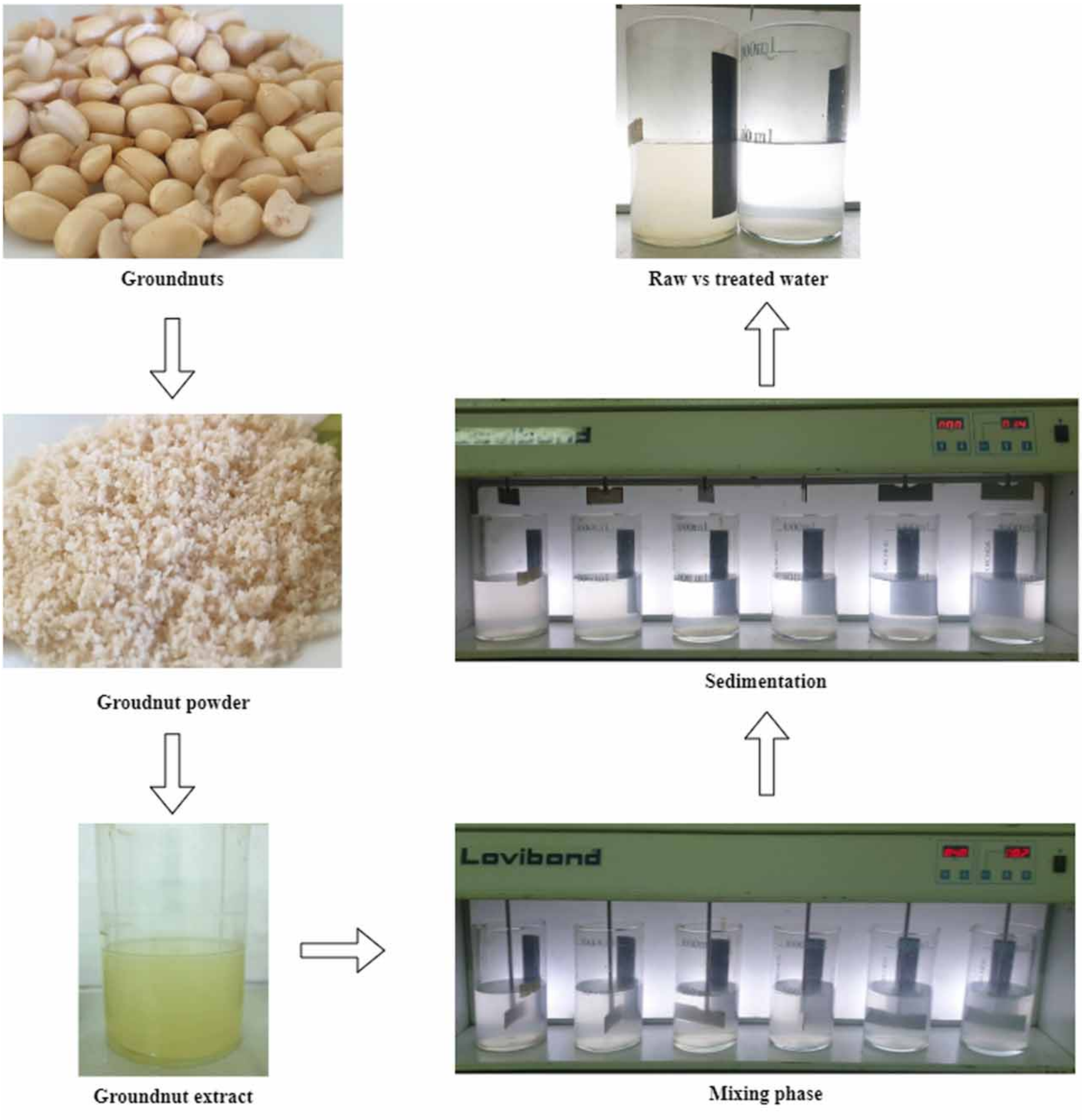
Natural coagulants can be an alternative solution to minimise the environmental pollution and health risks caused by the use of chemical coagulants. The main objective of this research was to evaluate the performance of groundnut extract for turbidity removal from water. For this, the active coagulant agents were extracted from the deshelled nuts and used in a series of water treatment tests performed at low, medium, and high turbidity levels using synthetic turbid water. The groundnut extract showed good coagulative abilities, close to those of $Al_2(SO_4)_3$. The best performance was obtained at medium turbidity (150 NTU), where the extract removed 89% of the turbidity using a 500 mg/L dosage. However, it was not as efficient as $Al_2(SO_4)_3$, whose coagulation was better at every turbidity level. The use of the groundnut extract does not modify the water's pH significantly and the floc size decreases as turbidity increases, although they are bigger than those produced by $Al_2(SO_4)_3$. Its flocculation and sedimentation processes are also quick (each less than 10 min) and quicker than those with $Al_2(SO_4)_3$. Hence, the groundnut extract is a viable alternative to chemical coagulants.

Key words: aluminium sulphate, groundnuts, natural coagulant, synthetic turbid water, turbidity removal, water treatment

HIGHLIGHTS

- Groundnut extract can be used efficiently to treat water at any turbidity level.
- Its application does not modify the pH of water significantly.
- The treatment process is quick, less than 15 min.

GRAPHICAL ABSTRACT



INTRODUCTION

Several treatment technologies are used to process raw water sources into drinking water. Most cannot be separated from the coagulation and flocculation stages. The coagulation–flocculation process is prevalent in water treatment due to its effectiveness in removing organic matter, suspended solids, turbidity, and colour (Abidin *et al.* 2013; Kakoi *et al.* 2016). The process requires the addition of positively charged, divalent chemical compounds. Negatively charged polymers are also used commonly in water treatment, notably as high molecular weight flocculants (Lapointe & Barbeau 2020).

The application of these compounds is not necessarily free from impacts (Barakwan *et al.* 2019b). The environmental impacts include increasing the corrosion rate of metallic utilities, changing the pH, limiting root elongation, and inhibiting seed germination. Water treatment involving conventional coagulants also generates excess chemical sludge in addition to the suspended solids to be removed; and this becomes another issue to resolve (Kurniawan *et al.* 2020). Aside from these impacts on the environment, concerns related to human health arise. Metal-based coagulants are non-biodegradable, and their residuals in drinking water can induce direct human health impacts when consumed and can accumulate in body cells (Kluczka *et al.* 2017). There have been reports of signs that chemical coagulants may have negative effects on human health, including central nervous system failure, dementia, Alzheimer's disease, and severe trembling (Ahmad *et al.* 2016; Exley 2017; Mirza *et al.* 2017; Barakwan *et al.* 2019a).

Natural coagulants may offer an alternative strategy to reduce the hazards to human health and the environment posed by chemical coagulants. They are entirely organic and biodegradable, and derived from living things; as a result, they are safe for the environment, with minimal potential negative effects on human health. However, a number of issues must be taken into account when using natural coagulants, mostly because of the complex extraction and purification processes, the limited availability of raw materials for continuous supply, and the varying characteristics of water and wastewater to be treated. The limitations of using natural coagulants open up new challenges for future study (Kurniawan *et al.* 2020). A lot of research related to natural coagulants is documented, but the findings are mainly focused on commonly known and established plants such as *Moringa oleifera*, *Jatropha curcas*, and *Opuntia ficus indica*. Although these sources have proven their ability to remove various pollutants, it is crucial to explore the possibility and feasibility of using other local raw material sources as well (Bahrodin *et al.* 2021).

This study's main objective was to evaluate the performance of groundnut extract in removing turbidity from water. Groundnuts or peanuts, taxonomically classified as *Arachis hypogaea*, are a legume crop of the Fabaceae family. It is known that peanut seeds contain arachin and coarachin, which are globulin proteins (Kristianto 2017). Proteins are proven active coagulant agents that work by charge neutralisation (Prihatinningtyas 2019). The focus of this study was on the development of a simple and reliable extraction method for the active coagulant agents contained in groundnuts, and to provide basic treatment process data, namely removal efficiency, change in pH, floc size, and flocculation and sedimentation durations. The effect of the water's initial turbidity was also determined.

MATERIALS AND METHODS

Preparation of groundnut extract

Deshelled groundnuts were purchased from the local market, washed with water, and sun-dried. They were then ground and 5 g of the finely broken groundnuts were inserted in 500 ml of 1 M NaOH (which was prepared by dissolving 40 g of sodium hydroxide pearls in 1 l of distilled water). The mixture was stirred at 200 rpm for 15 min to extract the coagulation active components. It was then allowed to sediment, and the liquid portion was collected and used as a coagulant (Figure 1).

Figure 1 | Deshelled groundnuts, finely broken groundnuts, and groundnut extract.

NaOH was chosen as the extraction solvent for the active coagulant agents (proteins) because it is efficient in protein extraction, and it is more readily available and affordable than other chemical solvents. Water and sodium chloride (NaCl) extracts were tested previously but showed poor results. This is in line with the work by Muda *et al.* (2020), who tested the same extraction solvents and concluded that NaOH was the most efficient.

Preparation of synthetic turbid water

The experiments aimed at evaluating not only the performance of the natural coagulant but also the effect of the water's initial turbidity. Synthetic turbid water was used for the experiments to enable control of the turbidity level while keeping other water characteristics constant.

Synthetic turbid water for treatment tests was prepared by adding kaolin to tap water. Kaolin was ground and sieved, and the finer fraction was used to prepare the turbid water. A 1% kaolin stock solution was made by adding 10 g of kaolin to 1 l of water. The solution was stirred for 1 h at 200 rpm, to achieve uniform kaolin dispersion, and then allowed to rest for 24 h for complete particle hydration (Figure 2). Immediately before the water treatment tests, synthetic turbid water samples with known initial turbidities were produced by diluting the stock solution with tap water. Three levels of turbidity were considered, as explained by Prihatinningtyas (2019):

Figure 2 | Preparation of synthetic turbid water.

- Low turbidity (T1): 50 NTU
- Medium turbidity (T2): 150 NTU
- High turbidity (T3): 300 NTU

Water treatment tests

The performance of the groundnut extract was determined using jar tests. Five hundred mL ml aliquots of synthetic turbid water of known turbidity were put into 1 l beakers. Various doses of the groundnut extract were then added to them and they were mixed at 110 rpm for 3 min initially, then at 40 rpm for 20 min. The water was then allowed to sediment for 15 min. Clarified samples were collected from the tops of the beakers, and the residual turbidity (T_s) was measured. The procedure was repeated while varying both the water's turbidity and the coagulant dosages. The same jar tests were performed without using a coagulant to represent the blank. The residual turbidity in the blank was noted as T_b. The removal efficiency was then calculated using the following equation

$$\text{Removal efficiency (\%)} = \frac{T_s - T_b}{T_b} \times 100 \tag{1}$$

The effect of using the natural coagulant on the water's pH was determined by measuring the pH of the water samples before and after each jar test. The evolution of floc formation was also observed and timed during flocculation in each jar test, as was the time taken for the majority of the floc formed to sediment. The whole experiment was repeated using aluminium sulphate as a coagulant.

Figure 3 is a schematic of the factorial experimental design. The green and blue blocks correspond to the natural (C1) and chemical (C2) coagulants, respectively. Vertical variations indicate changes in initial turbidity levels while coagulant dosages vary horizontally. D0 corresponds to the blank.

C1T1D0	C1T1D1	C1T1D2	C1T1D3	C1T1D4	C1T1D5
C1T2D0	C1T2D1	C1T2D2	C1T2D3	C1T2D4	C1T2D5
C1T3D0	C1T3D1	C1T3D2	C1T3D3	C1T3D4	C1T3D5
C2T1D0	C2T1D1	C2T1D2	C2T1D3	C2T1D4	C2T1D5
C2T2D0	C2T2D1	C2T2D2	C2T2D3	C2T2D4	C2T2D5
C2T3D0	C2T3D1	C2T3D2	C2T3D3	C2T3D4	C2T3D5

Figure 3 | Experimental design.

The chemical coagulant was used for the jar tests in five dosages (Table 1), for ease of comparison with the performance of the natural coagulant. The groundnut extract dosages are given in Table 2.

Table 1 | Tested dosages of aluminium sulphate

N°	Dosage (ml/0.5 l)	Dosage (ml/l)	Dosage (mg/L)
1	0.5	1	10
2	1.0	2	20
3	1.5	3	30
4	2.0	4	40
5	2.5	5	50

Table 2 | Tested dosages of the groundnut extract

N°	Dosage (ml/0.5 l)	Dosage (ml/l)	Dosage (mg/L)
1	5	10	100
2	10	20	200
3	15	30	300
4	20	40	400
5	25	50	500

RESULTS AND DISCUSSION

Effect of coagulant dosage and initial turbidity on removal efficiency

Using aluminium sulphate, the highest removal efficiency (97%) was observed at T3, with a dosage of 1 ml (Figure 4). T2 followed with 92% at 0.5 ml and T1 with 91% at 1.5 ml. As expected, aluminium sulphate removed more than 90% of the turbidity at every level.

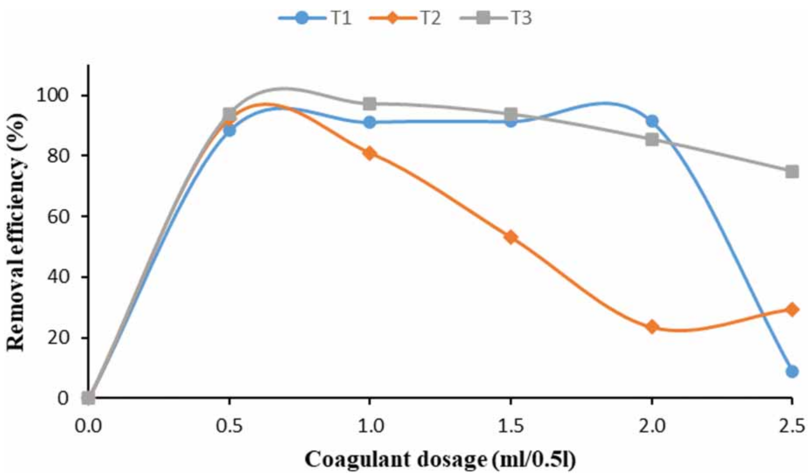


Figure 4 | Removal efficiency of aluminium sulphate.

The groundnut extract's best performance was at medium turbidity (Figure 5). Indeed, the removal efficiency at T2 was 89% with 25 ml as the optimum dosage. T3 efficiency was 87% at the same dosage. It is possible that an increase in dosage leads to a higher removal efficiency at this level. The optimum dosage of 10 ml led only to a

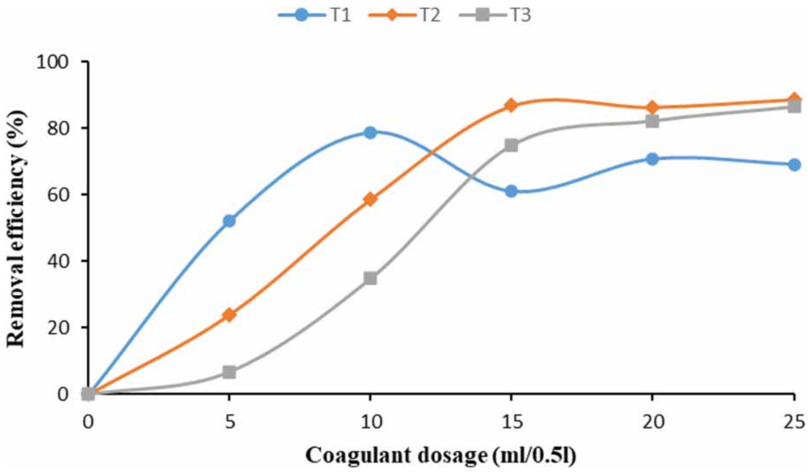


Figure 5 | Removal efficiency of groundnut extract.

removal efficiency of 79% for T1. While this is appreciable, groundnut extract cannot provide the turbidity removal performance of $Al_2(SO_4)_3$. Figure 6 shows water samples before and after treatment using groundnut extract.

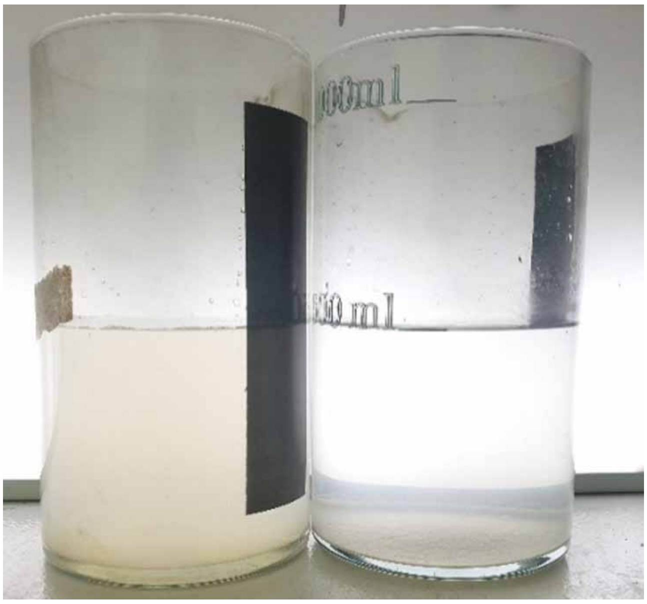


Figure 6 | Untreated (left) and treated (right) water.

Some studies, though with different protocols, have also demonstrated that groundnut extract can remove turbidity successfully from water. Raed *et al.* (2023) opted for 1 M NaCl extraction of de-oiled peanut seeds and achieved 75.4–85.0% turbidity removal from water samples with 35.2 NTU mean turbidity, while alum exceeded 90.0%. Prasad *et al.* (2017) achieved 84% turbidity removal from a 100 NTU water sample using a water-based groundnut seed extract dosed at 20 mg/L. Abood *et al.* (2017) treated a 340 NTU water sample using a 4 mol/l KCl groundnut extract and obtained 88.3% turbidity removal efficiency. Birima *et al.* (2013) reported that peanut (after oil removal) extracted with 6 mol/l NaCl solution could effectively remove 92% of the 200 NTU turbidity using 20 mg/L, while peanut seeds extracted with distilled water could remove only 31.5% of the same turbidity with the same dosage and those extracted with 1 M NaCl removed about 40%. In our experiments, however, water-based groundnut extract was not able to induce coagulation in water and 1 M NaCl did not give significant results, hence the choice of 1 M NaOH as extraction solvent. It is possible that increasing the dosage of NaOH increases the turbidity removal efficiency of groundnut extract. Removing the oil from groundnuts before extraction is also likely to increase our extract's performance. Half of the dry weight of peanut seeds consists of lipid fractions that do not contribute to their removal efficiency. Therefore, these lipidic fractions should be removed to enhance the coagulation power of peanut seed cake (Choy *et al.* 2015). This explains the high coagulant dosages that were required for our treatment.

Effect of coagulant dosage and initial turbidity on pH

As regards the water's pH, the use of the aluminium sulphate induced almost no pH change (Figure 7). At T1, the pH was constant (7.00) for every dosage. At T2, it was constant at 6.99 but the pH of the blank was 7.00. This means that the coagulant caused a change in pH of 0.01. T3 behaved differently. The addition of 0.5, 1 and 1.5 ml dosages had no effect on the pH, but higher dosages lowered it by 0.01. In general, $Al_2(SO_4)_3$ had no significant effect on the water's pH.

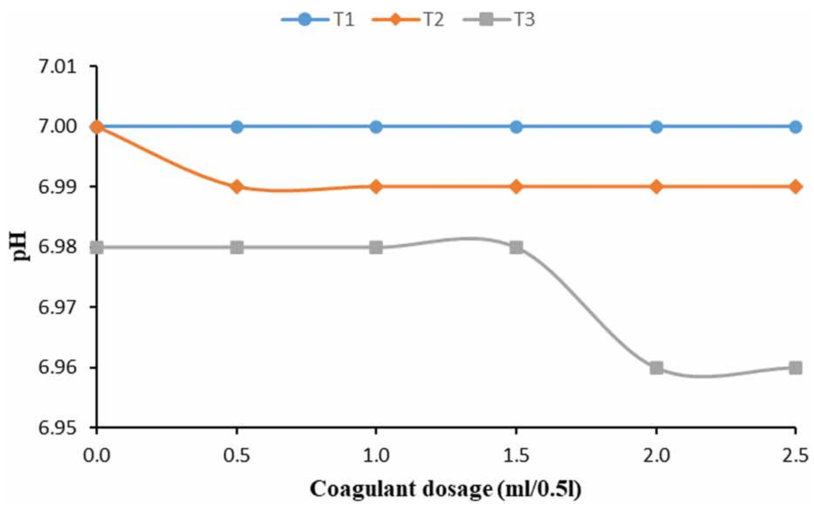


Figure 7 | Water pH after treatment using aluminium sulphate.

The use of groundnut extract also induced almost no pH change in the water, as shown in Figure 8. At T3, the pH was constant (7.00) for all dosages. At T2, there was a pH change of 0.01, which is not significant. T1 on the other hand, had more but equally small variations. The addition of 5 and 10 ml aliquots did not affect the pH, but higher dosages increased it by 0.01. Thus, if the water to be treated already has the required pH, it will not be modified upon clarification using groundnut extract. Even though the coagulant is basic due to NaOH, the basicity is removed from the water along with the sediments after treatment.

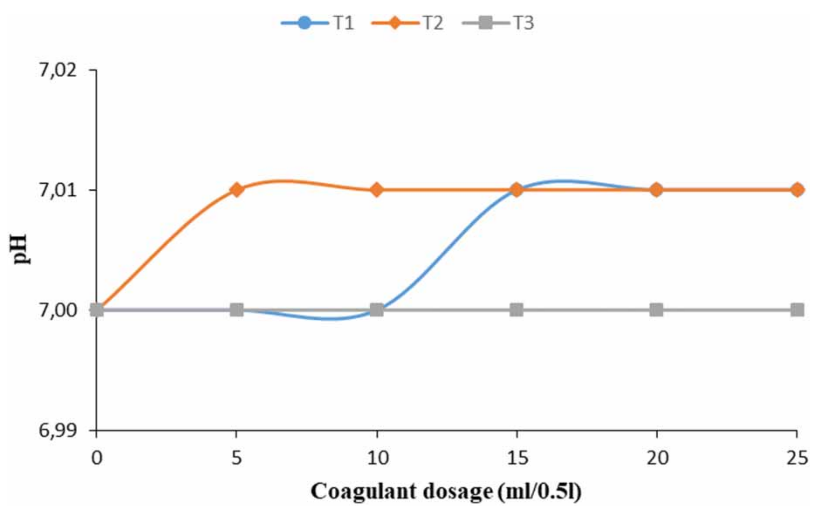


Figure 8 | Water pH after treatment with groundnut extract.

This water pH conservation is consistent with that reported by Prasad *et al.* (2017), who observed a pH change from 8.3 to 8.4 at the optimum dosage. Abood *et al.* (2017) also reported only a small pH change from 7.5 to 7.3 at optimum dosage.

Effect of coagulant dosage and initial turbidity on flocculation time

Table 3 details the time taken for flocculation with $Al_2(SO_4)_3$ to appear complete at each dosage and turbidity level. The flocculation time is constant (7 min) for every dosage at T1, except 2.5 ml where it is 20 min, the duration of the mixing phase. At T2, more water samples flocculated extremely slowly, except for dosages 0.5 and 1 ml, where the flocculation time was 9 min. At T3, 7 min was enough for dosages between 0.5 and 1.5 ml, and 10 min for higher dosages. The water samples which flocculated slowly are those in which turbidity removal was low.

Table 3 | Flocculation time using aluminium sulphate

	Flocculation time		
Dosage (ml/0.5 l)	**T1**	**T2**	**T3**
0.5	7	9	7
1.0	7	9	7
1.5	7	20	7
2.0	7	20	10
2.5	20	20	10

Table 4 details the time taken for flocculation with groundnut extract to appear complete at each dosage and turbidity level. At dosages between 10 and 25 ml, the flocs formed quickly whereas the 5 ml dosage always had the longest flocculation time. At 5 ml, half the mixing duration had passed before a significant number of flocs could be observed in the water, hence the low turbidity removal. Overall, flocculation is quicker with groundnut extract than with $Al_2(SO_4)_3$ at optimum dosages and it can be concluded that it is time-saving to use groundnut extract rather than $Al_2(SO_4)_3$ for water treatment. The total duration of the mixing phase can be reduced to 10 min or less for low turbidity water and 5 min for medium and high turbidity waters.

Table 4 | Flocculation time using groundnut extract

	Flocculation time		
Dosage (ml/0.5 l)	**T1**	**T2**	**T3**
5	10	17	8
10	6	6	6
15	6	4	6
20	4	4	4
25	4	3	4

Effect of coagulant dosage and initial turbidity on floc size

Table 5 details the sizes of the flocs formed at each dosage and turbidity level using $Al_2(SO_4)_3$. The flocs formed were small at both T2 and T3 for smaller dosages, and very small for higher dosages. At T1, from 1 to 2 ml, the flocs were of medium size. The higher dosage resulted in the formation of very small flocs while the lower dosage resulted in the formation of small flocs.

Table 6 details the sizes of the flocs formed at each dosage and turbidity level, using groundnut extract. Big flocs were formed at T1 and T2 at optimum dosages of 10 and 25 ml, respectively, while the 5 ml dosage had the smallest floc sizes. At T1, medium sizes were observed at higher dosages. At T2 and T3, however, floc size

Table 5 | Floc size using aluminium sulphate

Dosage (ml/0.5 l)	Floc size		
	T1	T2	T3
0.5	S	S	S
1.0	M	S	S
1.5	M	VS	S
2.0	M	VS	VS
2.5	VS	VS	VS

M, medium; S, small; VS, very small.

Table 6 | Floc sizes using groundnut extract

Dosage (ml/0.5 l)	Floc size		
	T1	T2	T3
5	S	VS	VS
10	B	S	VS
15	M	S	S
20	M	M	S
25	M	B	S

VS, very small; S, small; M, medium; B, big.

increased with dosage. This is in accordance with the flocculation times and coagulation activities that were reported. Overall, groundnut extract produced bigger flocs than $Al_2(SO_4)_3$ and does not require an additional product to aid with flocculation.

Effect of coagulant dosage and initial turbidity on sedimentation time

Table 7 details the time taken for more than 90% of $Al_2(SO_4)_3$ flocs to sink to the bottom of the beaker at each dosage and turbidity level. At T1, sedimentation was quick (less than 5 min), except for the 2.5 ml dosage where the sedimentation time allowed (15 min) was not enough. The 15-min duration was also insufficient for the three last dosages at T2. The sedimentation time was medium at 0.5 and 1 ml at this turbidity level. 8 and 10 min were obtained, respectively. At T3, there were variations ranging between 4 and 8 min for dosages 0.5–1.5 ml. A correlation can be seen between sedimentation time and removal efficiency.

Table 7 | Sedimentation time using aluminium sulphate

Dosage (ml/0.5 l)	Sedimentation time		
	T1	T2	T3
0.5	4	8	5
1.0	3	10	4
1.5	3	>15	8
2.0	3	>15	15
2.5	>15	>15	15

Table 8 details the time taken for more than 90% of groundnut extract flocs to sink to the bottom of the beakers at each dosage and turbidity level. Sedimentation time increased as turbidity increased. It was between 3 and 5 min for T1 and T2, but got higher at T3, to the point where most flocs were still in suspension when the allowed sedimentation time was over for lower dosages. This obviously resulted in lower coagulation activities. A better

Table 8 | Sedimentation time using groundnut extract

	Sedimentation time		
Dosage (ml/0.5 l)	**T1**	**T2**	**T3**
5	3	5	>15
10	3	5	>15
15	3	5	14
20	5	4	10
25	5	3	8

performance can be expected if the sedimentation time is prolonged. Sedimentation is quicker with groundnut extract than with $Al_2(SO_4)_3$ due to the bigger floc sizes. The sedimentation duration in the water treatment protocol can be reduced to 5 min for low and medium turbidity waters, and 10 min for high turbidity waters.

Relationship between optimum dosage and initial turbidity

The optimum aluminium sulphate dosages for T1, T2, and T3 were 1.5 ml/0.5 l, 0.5 ml/0.5 l, and 1 ml/0.5 l respectively, all removing more than 90% of the turbidity in the water. For the groundnut extract, the optimum dosages for T1, T2, and T3 were 10 ml/0.5 l, 25 ml/0.5 l, and 25 ml/0.5 l, respectively. The relationship between optimum dosage and initial turbidity appears to be logarithmic, as shown in Figure 9, and a logarithmic model was derived, corresponding to the trend curve and equation shown in the figure. The coefficient of determination (R^2) is 0.8439, implying that the model is a good fit and can serve as the basis for future studies and an eventual implementation in a water treatment plant.

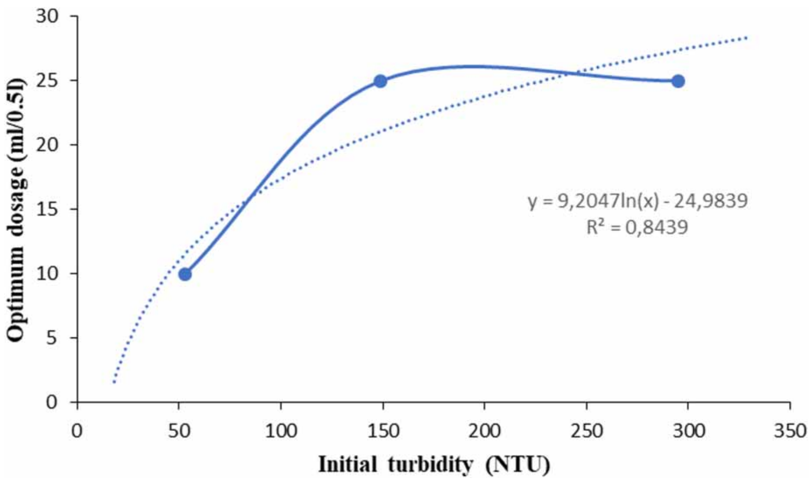


Figure 9 | Optimum dosages obtained using groundnut extract.

CONCLUSIONS

Groundnut extract is a promising natural coagulant for turbidity removal from water. The best performance was obtained at medium turbidity level (150 NTU), where the extract removed about 89% of the turbidity using a dosage of 500 mg/L. Sodium hydroxide is efficient in extracting the active coagulant agents from groundnuts. However, the extract was not as efficient as $Al_2(SO_4)_3$, whose coagulation was better at every turbidity level. The use of groundnut extract does not modify the pH of water significantly. The floc size decreases as turbidity increases but they are bigger than those produced by $Al_2(SO_4)_3$, and flocculation and sedimentation are quick (each less than 10 min), quicker than those with $Al_2(SO_4)_3$. Groundnut extract can be used efficiently to treat water with any turbidity level, the required dosage depending on the characteristics of the water to be treated. Groundnuts are readily available and environmentally friendly, both of which tend to facilitate remote area application as a source of natural coagulants. Hence, groundnut extract is a viable alternative to chemical coagulants.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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