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Research Article

An approach for analysis and selection of ideal natural coagulants for the treatment of synthetically prepared turbid water

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Abstract
The volume of research on natural coagulants has expanded significantly in recent years, driven by increased awareness of environmental sustainability and the excellent characteristics and efficiency that natural coagulants offer, thereby minimising environmental effects. The present study aimed to select the most suitable natural coagulants among banana peel, coconut fibre, groundnut shell, onion peel, sawdust, and lemon peel, necessitating a systematic approach, such as a screening test. The experimental runs were conducted using a standard jar test apparatus having synthetic turbid water to assess the efficiency of coagulants. The physicochemical properties were examined using the standard methods recommended by APHA (2017). Color, turbidity, pH, electrical conductivity, total solids, and sludge dewaterability were chosen as the primary indicators for this study. The results revealed encouraging insights into the efficacy of the tested natural coagulants, with rice husk removing the least amount of turbidity (83.3%) at 1gm/500ml and onion peel removing the most at 96.4%. The sludge formed after treatment with natural coagulants demonstrated outstanding dewaterability, with tamarind seeds having the lowest dewaterability at 29.17% and coconut fibre having the highest at 90.2% at a dosage of 1 gm/500 ml. The findings suggest that tested natural coagulants are effective for water treatment. Furthermore, sludge dewaterability, another critical measure in analysing feasibility and sustainability, was reported to be the greatest by coconut fibre (90.2%) and the least by onion peel at 46.71%.The screening strategy used in the study appears to be quite effective in expediting the selection process by systematically examining the influence of coagulant type and dose. The study not only identified the most effective coagulants but also saved time and effort by eliminating less effective choices.

Keywords: Coagulation, Natural coagulants, Screening, Sludge, Turbidity

INTRODUCTION

In the contemporary world, a wide spectrum of alternatives or solutions is available for various products, but it is the process of finding meaningful solutions that matters, which enables new experiences, inspires, and generates a positive impact on society and the environment. Therefore, sustainable products are looked by the global population (Al-Oqla and Sapuan, 2017). The selection of appropriate materials has a significant impact on research and development. Inappropriate selection of materials may result in the system's process/efficiency failure. Selecting materials that best provide the solution, give the best performance, and have the least cost is the goal of research. Over the last few decades, many traditional materials have been replaced by new materials, thereby increasing the existing set of supplies in terms of type and number. Due to the enormous number of materials and the multifaceted relationships among several selection parameters, material selection is usually a difficult and tedious task, making it more challenging than before. Several methods have been proposed previously to address the issue of material selection and enhance efficiency in material development. Some of the recent methods include the multiple decision-making method for the selection of materials. These methods emphasizes the importance of material selection (Kumar and Ray, 2014). To achieve this objective, the selection process of natural fibres becomes crucial for achieving efficient results (Wankhede *et al*., 2023).

The behaviour of materials is governed by principles that are grounded in science and understandable. The characteristics of materials are determined by their structure, which changes concerning the environmental conditions. When selecting material for a specific purpose, it is essential to conduct appropriate and sufficient testing to ensure that the material remains suitable for its intended use. Five steps have been proposed for material selection by Chiner (1998), which include design definition, material properties analysis, screening of materials, assessment and verification of the optimal solution, and tests for confirmation. Several others, such as Jalham (2006), Van Kesteren *et al.* (2006), Ashby (2004), and Farag (2002), have also proposed various steps for the selection of materials. Among various steps proposed by all the above the common and significant processes in material selection are screening and ranking. Accordingly, several quantitative methods have been developed toanalyse the materials for a systematic evaluation (Jahan *et al*., 2010).

Awareness among the population regarding the unexpected damage caused by synthetic materials has led to the development of eco-friendly materials. Furthermore, researchers have also demonstrated a strong interest in developing materials that can substitute for synthetic materials. This has led to an increase in demand for natural fibre-based composites for various applications in recent years. Biodegradable materials and natural fibres contribute to developing a “green” economy due to their merits, including ease of manufacturing, energy efficiency, eco-friendliness, economic benefits, and sustainability. Natural fibres have become increasingly appealing to research and innovation in recent years as alternatives to synthetic/chemical fibres due to the aforementioned qualities (Makinde-Isola *et al*., 2024).

Usually, the first treatment unit in the conventional water treatment process is coagulation and its efficiency directly affect all downstream treatment steps. The effectiveness of coagulants is determined by the formation of polynuclear molecules, which are multicharged and possess adsorption capability after hydrolysis. In this concern Bratby (2016) stated that the charges formed is directly proportional to the coagulation process (Bratby, 2016). The ratio of sludge produced to the addition of coagulant is calculated as the yield and acts a criterion for the effectiveness of coagulants. The lower the ratio of the sludge produced against the coagulant used, the more efficient the coagulant will be (Hamawand *et al*., 2017).

Natural coagulants have demonstrated reliable performance in water and wastewater treatment, effectively removing most pollutant parameters. Previous studies have shown that the efficiency of natural coagulants in comparison with metal-based coagulants is undeniably great, and these can achieve similar or even superior performance in pollutant removal. A literature review states that most research on natural coagulants originates from developing and tropical countries, which can be attributed to the availability of diverse and abundant resources (either from waste or by-products) (Adnan *et al*., 2017; Kristianto, 2017; Antov *et al*., 2018). Therefore, plant-based coagulants still receive special attention in this research area. Nevertheless, research on utilising waste or by-products as coagulants is still limited, and further study in this area could be a future direction (Othman *et al*., 2018; Zaidi *et al*., 2019). The objective of the present work was to select suitable coagulants for treating synthetically prepared turbid water.

MATERIALS AND METHODS

Materials

Natural coagulants were collected from the source of production. The present study used six coagulants: rice husk, sawdust, lemon peel, tamarind seeds, coconut fiber, and onion peel. Rice husk, tamarind seeds, and coconut fiber were collected from rural areas near Visakhapatnam District. At the same time, sawdust was obtained from a wooden furniture manufacturing unit in the city's outskirts, and lemon peels and onion peels were collected from local eateries. Chemicals of analytical reagent grade were used to analyse water samples before and after coagulation, as well as to prepare turbid water (Kaolin).

Methods

Preparation of coagulants

The coagulants were cleaned with tap water, oven-dried, then ground into a fine powder using a blender. The powders were stored in airtight vials at room temperature for future usage (Dwarapureddi *et al*., 2021).

Preparation of synthetic turbid water

A 10g kaolin stock solution was mixed with one litre of distilled water to make synthetic turbid water. 10 mL of the stock solution was dissolved in 500 mL of water to yield a 150 NTU solution. The initial concentrations of pH, turbidity, conductivity, color, and TDS are given in Table 1.

Water analysis

The study evaluated the following water quality measures i.e. colour, turbidity, pH, electrical conductivity, total dissolved solids and sludge weight pre and post-coagulation, which were analyzed following the standard methods (APHA, 2017)

Table 1. Initial values of parameters of the synthetic turbid water

Parameters	Initial values (Mean ±SD)
pH	7.45 (7.23±0.17)
Turbidity (NTU)	150 (137±0.10)
Conductivity (µs/cm)	0.6 (0.56±0.04)
Colour (Pt-Co Scale)	0.17 (1.15±0.03)
TDS (mg/L)	214 (204±0.29)

Range mentioned in parentheses

Coagulation experiments

The effectiveness of the coagulants under investigation was evaluated using a traditional Jar test device. Each of the four beakers held 500 ml. of kaolin suspension. Coagulants in the following 4 doses: 1 g, 2 g, 3 g, and 4 g were added. After that, the samples were quickly combined for 2 minutes at 80 rpm, then slowly mixed for 30 minutes at 30 rpm. After that, the samples were maintained in a quiescent state for thirty minutes. Filtered specimens were obtained for the study of the post -coagulants. The following formula was used to calculate the percentage decrease in turbidity.

$$Turbidity\ Reduction\ (\%) = \frac{Initial\ Turbidity - Final\ Turbidity}{Initial\ Turbidity}\ X\ 100 \quad \text{Eq.1}$$

Wet and dry sludge

Wet sludge is the weight of sludge recovered from the filtering water sample after coagulation. This wet sludge was oven-dried for 6 to 8 hours at 150 $^{\circ C}$ to remove its moisture content, and the resulting product is the dry sludge.

Statistical analysis

The effect of coagulant concentration (ppm) on turbidity removal percentage and sludge formation was determined using analysis of variance (ANOVA),using SPSS software

RESULTS AND DISCUSSION

Selection of coagulants

Material selection was the process of selecting the finest material for a specific purpose through an organised material selection approach. The process was adopted as a preliminary screening for testing the efficiency of coagulants. The screening process helps eliminate materials that do not satisfy the requirements (Sotoodeh, 2018). Selecting a coagulant is not easy, as it can effectively remove suspended solids, but it may also increase conductivity. This makes the final selection of coagulant dependent on the relative importance of each measured parameter (Tzfati *et al*., 2011). In the present study, the selection of an optimal material was based on two parameters: turbidity reduction and generated sludge, in addition to considering changes in pH, reduction in colour, electrical conductivity, and total dissolved solids.

After selecting natural coagulants, screening was carried out to treat sludge with coagulant doses and physicochemical parameters such as turbidity, pH, conductivity, color, and TDS.

The pH changes after the treatment with natural coagulants arepresented in Fig. 1. Lemon peel showed a significant decrease from 7.3 to 4.53 followed by onion peel from 7.01 to 9, while the pH changes by other coagulants (banana peel, coconut fiber, groundnut shell, onion peel and saw dust) were meagre (Table 1).

Coagulants and pH of the treated water

The pH changes after treatment with the tested coagulants are shown in Fig. 1. The tested coagulants exhibited different trends in the pH of treated water; while banana peel, lemon peel, and coconut fibre reduced the pH, groundnut shell, onion peel, and sawdust enhanced the pH of treated water. Although the changes, i.e., reduction and increase of pH, are not significant except for lemon peel, which decreased pH from 7.3 to 4.5 at a dose of 4g/500ml. This can be attributed to the existence of citric acid, a tricarboxylic organic acid found in the peel of citrus (Li *et al*., 2019; Capanoglu *et al*., 2023).When alum is used as a coagulant, the pH of the treated water is reduced from 7.6 to 4.2 (Ndabigengesere and Narasiah, 1998), which is due to the hydrolysis process that does not occur with polymeric coagulants. Studies where natural coagulants, such as Moringa oleifera, are used as coagulants have not presented any noteworthy changes in the pH of the treated water (Cunha *et al.,* 2023) (Table 2).

Furthermore, Skoronski *et al*. (2017) reported that when a coagulant is added to water, it is essential to ensure that it does not enhance the concentration of dissolved substances, which could, among other effects, cause a significant change in the pH of the treated water, resulting in difficulty with sedimentation. One reason the tested coagulants did not show a significant pH change is due to the lower doses used. When *M.oleifera* was used as a coagulant and scallop shell powder as an antibacterial agent (Zaman *et al*., 2017) for treating water followed by biofiltration, the treated pH of the water ranged from 7.37 to 7.89 (initially 7.62 to 8.01), which is similar to the observations in the present study.

Fig. 2 illustrates the removal of conductivity achieved through the use of natural coagulants. Conductivity was induced after treatment with coagulants such as banana peel, onion peel, and coconut fibre, while coagulants such as groundnut shells and sawdust did not change. The highest induced conductivity was observed with banana peel (-73.33%) at a dose of 4 mg/500 ml, while the lowest conductivity induced (-16.66%) was observed with coagulants, including coconut fibre, onion peel, and lemon peel. Groundnut and sawdust have did not show any change (0%) (Table 2).

Coagulants and electrical conductivity

Four of the tested coagulants induced conductivity, with the highest being Onion peel at 4g/500ml, resulting in a decrease of -73.33%, and the least induced being -16.66% by all the coagulants at various doses. Studies by El Mouhri *et al*. (2024) showed that the conductivity removal amounts were low, apparently because the effluent's electric phase is dominated by ions with a similar charge to colloids, leading to repulsive forces. Similarly, as the dose of coagulant increased, the total number of counterions also increased, enhancing the probability of oppositely charged molecules agglomerating with the polymer after neutralisation (Neffa *et al*., 2020). The coagulants saw dust and groundnut shells have not induced any conductivity in the present study.

Figure 3 depicts turbidity, total dissolved solids, and color removal using natural coagulants. The highest turbidity removal was obtained with onion peel (96.4%) and the lowest was obtained with banana peel (83.33%). TDS was induced after treatment by all the coagulants, and the color removal was reported to be very meagre by all the coagulants (Table 1).

Coagulants and removal of turbidity, total dissolved solids and colour

The tested coagulants have shown various trends in turbidity reduction. The highest turbidity removal was achieved by onion peel (96.40%) at a concentration of 1g/500ml, and the least was recorded by banana peel (83.33%) at the same concentration. Coagulants such as banana peel, groundnut shell, lemon peel, and sawdust showed a turbidity removal that was directly proportional to the increasing dose. In contrast, coconut fibre and onion peel showed turbidity removal that was inversely proportional to the increasing dose. Turbidity reduction, directly proportional to the coagulant dose, is in line with the results obtained by Ahmad *et al*. (2022) when P. sarmentosum was used as the coagulant. Turbidity removal of up to 90.2% was reported in previous studies that used plant leaves (*Pandanus, Centella asiatica, and Cymbopogon citratus* leaves) as coagulants, adopting different extraction methods using water, sodium chloride, and sodium hydroxide (Muda *et al*., 2020). The present study achieved higher (87 – 96%) turbidity reduction without any extraction methods and using coagulant in the raw powdered form. Previous studies have shown that rice husk can be used as an aqueous coagulant aid to enhance the effectiveness of chemical coagulants, as it is rich in silica and can form larger and denser flocs (Tan *et al*., 2022).

All the tested coagulants induced total dissolved solids with increasing doses. Banana peel at 4g/500ml induced the highest decrease (-75.68%), while the least was recorded by groundnut shell at 1g/500ml (-0.48%).Ali (2017) presented that the biomass of banana peel contains chemical groups, such as hydroxyl groups, phosphates, and carboxylic acids, which act as active sites for the adsorption and removal of dissolved solids, chemical oxygen demand, and colour from water and wastewater (Azamzam *et al*., 2022). A 16.07% and 24.18% reduction in total dissolved solids was obtained when sawdust was used as an adsorbent without and with washing, respectively (Al-Tayyar and Najm, 2023).

Colour removal by all the tested coagulants was directly proportional to the coagulant dose. Nevertheless, it was very low, with a maximum of 0.07%, at a 4g/500ml

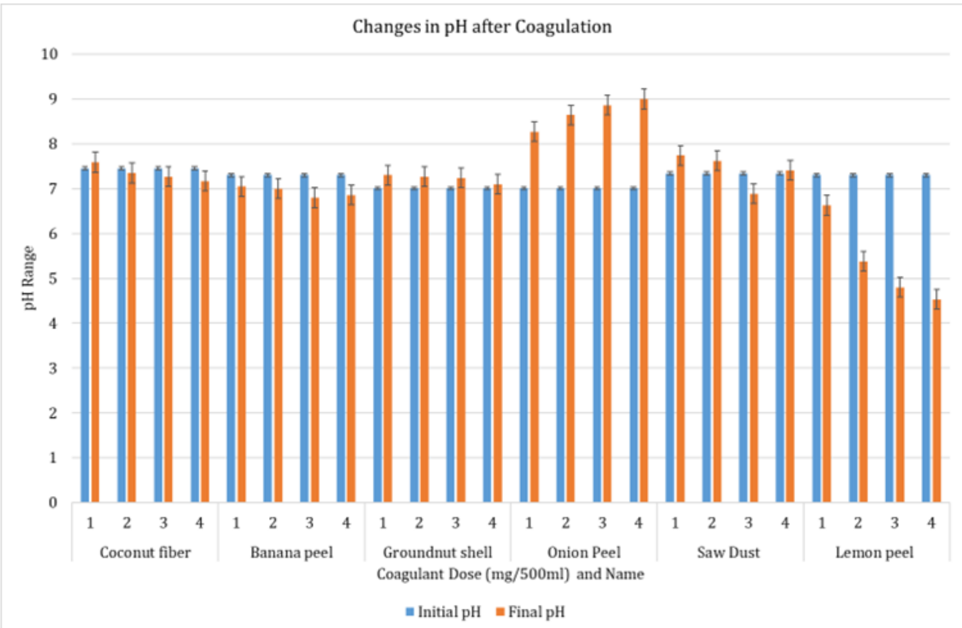

Fig. 1. *Changes in pH of synthetic turbid water after treatment with coagulants*

Table 2. Values/per cent change of various parameters after treating with natural coagulants

Name of the Coagulant	Dose of Coagulant (gms)	Mean ±SD (Percent removal given in parenthesis)			
		Conductivity (µs/cm)	**Turbidity (NTU)**	**Total Dissolved Solids (mg/L)**	**Colour (Pt-Co Scale)**
Coconut fiber	1	0.71±0.01 (-16.66)	8.37±0.04 (+94.4)	254.50±2.12 (-19.62)	0.43±0.00 (+0.02)
	2	0.79±0.01 (-33.33)	7.49±0.01 (+95)	289.00±1.41 (-35.51)	0.7 (0.79±0.01) (+0.04)
	3	0.89±0.01 (-50)	14.69±0.01 (+90.2)	324.50±2.12 (-52.33)	1.17±0.00 (+0.06)
	4	1.05±0.07 (-66.66)	18.50±0.07 (+87.33)	360.00±2.83 (-69.15)	1.49±0.00 (+0.07)
Banana peel	1	0.83±0.04 (-33.33)	24.00±1.41 (+83.33)	305.00±2.83 (-40.11)	7.65±0.08 (+0.03)
	2	1.05±0.07 (-53.33)	18.90±0.14 (+87,33)	389.00±2.83 (-78.6)	0.46±0.02 (+0.03)
	3	1.35±0.07 (-63.33)	17.90±0.14 (+88.13)	491.65±2.12 (-98.6)	0.65±0.01 (+0.05)
	4	1.68±0.04 (-73.33)	14.20±0.42 (+90.73)	459.50±0.71 (-100)	1.07±0.01 (+0.06)
Groundnut shell	1	0.59±0.01 (0)	12.75±0.35 (+91.66)	224.00±1.41 (-0.48)	0.14±0.00 (+0.01)
	2	0.61±0.01 (0)	13.00±0.28 (+91.46)	240.50±2.12 (-7.65)	0.24±0.00 (0.01)
	3	0.60±0.01 (0)	12.28±0.35 (+91.6)	250.50±2.12 (-12.16)	0.42±0.01 (+0.02)
	4	0.70±0.01 (-16.66)	11.90±0.14 (+92.13)	250.50±2.12 (-18.91)	0.50±0.01 (+0.03)
Onion Peel	1	0.71±0.01 (-16.33)	5.60±0.28 (+96.4)	258.00±2.83 (-15.13)	0.00±0.01 (+0.00)
	2	0.81±0.01 (-33.33)	7.45±0.21 (+95.13)	287.00±2.83 (-28.13)	0.02±0.00 (+0.00)
	3	0.80±0.01 (-33.33)	9.05±0.21 (+94.06)	308.50±2.12 (-38.22)	0.05±0.00 (+0.01)
	4	0.91±0.01 (-50)	12.60±0.57 (+91.86)	336.00±1.41 (-50.9)	0.07±0.00 (+0.01)
Saw Dust	1	0.61±0.01 (0)	16.00±0.28 (+89.46)	230.00±2.83 (-5.04)	0.08±0.00 (+0.01)
	2	0.60±0.01 (0)	12.70±0.42 (+91.76)	234.00±1.41 (-6.42)	0.11±0.01 (+0.01)
	3	0.59±0.02 (0)	11.95±0.64 (+92.33)	244.00±2.83 (-11)	0.12±0.00 (+0.01)
	4	0.59±0.01 (0)	10.00±0.28 (+93.46)	242.50±2.12 (-10.55)	0.19±0.00 (+0.01)
Lemon peel	1	0.51±0.01 (-16.66)	22.80±0.28 (+84.33)	179.50±2.12 (+17.31)	- 0.44±0.01 (+0.02)
	2	0.62±0.02 (0)	18.00±0.28 (+88.13)	196.00±2.83 (+11.31)	0.77±0.01 (+0.04)
	3	0.61±0.01 (0)	19.15±0.49 (+87.46)	232.50±2.12 (-5.47)	1.03±0.00 (+0.05)
	4	0.71±0.01 (0)	19.10±0.99 (+87.73)	260.00±1.41 (-18.26)	1.37±0.05 (+0.07)

dose, using lemon peel and coconut fibre. When used as biosorbents for the removal of textile dyes, groundnut shells showed that the adsorption efficiency increased with a decrease in particle size, and the maximum adsorption obtained was 46.80 mg/g (Lazarova *et al*., 2023). Studies by Jakka *et al*. (2023) presented that the nano-cellulose derived from coconut coir was an effective nano-adsorbent for dye removal. Ahamad *et al*. (2023) claimed that sawdust from Azadirachta indica is a unique and effective adsorbent that can remove crystal violet dye even at lower doses.

Figure 4 depicts the weights of sludge formed following treatment with natural coagulants. The wet and dry weights of the sludge, along with the difference, are

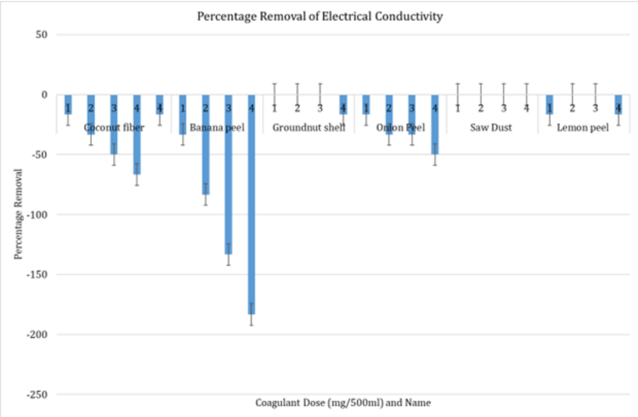

Fig. 2. *Percentage removal of electrical conductivity of synthetic turbid water by coagulants*

presented in the graph. The difference in sludge generated is high for coconut fibre (about 32.55g), followed by onion peel (28.97g), and the least is reported for banana peel (3.81g).

Sludge generated by coagulants

To achieve better performance, a thorough understanding of coagulant and pollutant reactions is necessary. The present study have noted that the sludge generated by the natural coagulants has dried sludge (3.81mg). Similar results were obtained from the previous studies. They have also stated that the sludge generated by natural coagulants is beneficial in many aspects, such as not adding any metals to the treated water, reducing sludge production and thus reducing disposal cost (Owodunni *et al*., 2021; Alazaiza *et al*., 2022; de Oliveira *et al*., 2021). Justina *et al*. (2018) presented evidence that sludge formed from tannin-based natural coagulants has the advantages of being biodegradable and poses no human health risks.

The turbidity removal and sludge dewaterability achieved by using natural coagulants are shown in Fig. 5. For coconut fiber, onion peel and sawdust, turbidity removal and sludge dewaterability decreased as the coagulant dose increased (94.4% at a dose of 1mg/500ml while 87.33% at a dose of 4mg/500ml), while for banana peel and groundnut shell, turbidity removal and sludge dewaterability increased as the coagulant dose increased (83.33% at 1mg/500ml and 88.13% at 4mg/500ml).

Turbidity was reduced after treatment with natural coagulants, indicating high degree of particle incorporation into the flocs. The flocs formed are likely a result of cluster-cluster aggregation. It is seen that the natural coagulants have generated smaller floc size. Additionally, natural coagulants are recognised for their high surface charge and short chain length. Generally, the flocs formed are fractal, implying that density reduces with increasing floc size (Gregory, 1997), which is a result of enhanced fragmentation and erosion of larger flocs, causing the release of smaller particles into the solution and increasing residual turbidity (Haasler *et al*., 2023). Sludge dewatering depends on the type of floc formed. For example, if the floc are very young, which are formed at high loads or due to severe changes or disturbances, then the residual turbidity will be high owing to greatly dispersed growth (Ericsson 1988). The correlation between sludge volume ratio and residual turbidity can be inverse at times, when irregular and/or filamentous structures are able to sweep fine material during settling (Sezgin *et al*., 1978).

On the other hand, floc strength refers to the capability to retain good filterability after exposure to high shear.

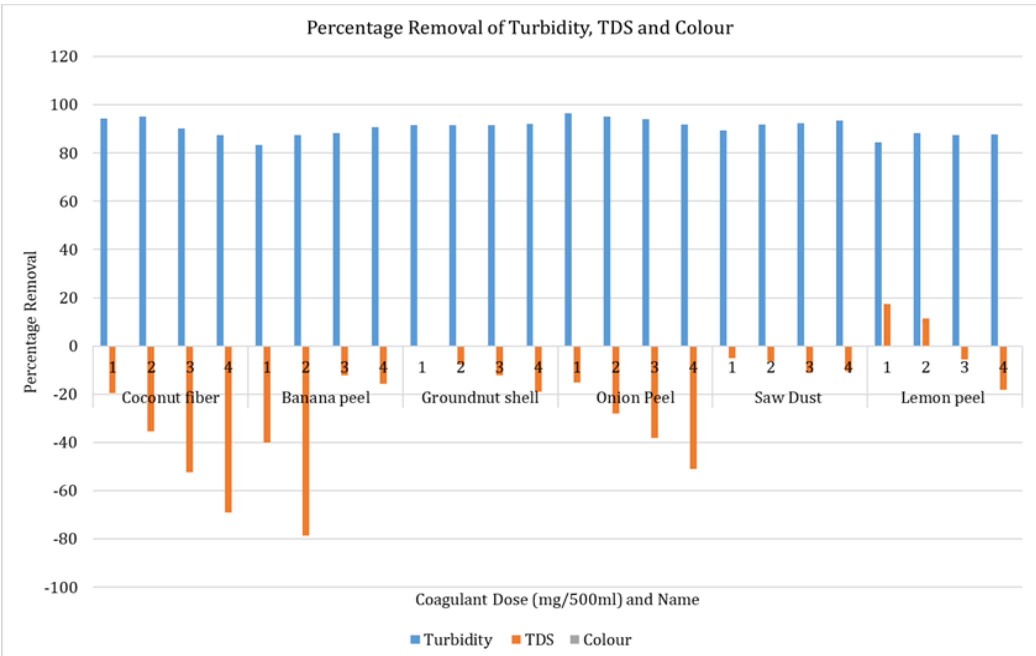

Fig. 4. *Wet and dry weights of the sludge generated from synthetic turbid water after treatment*

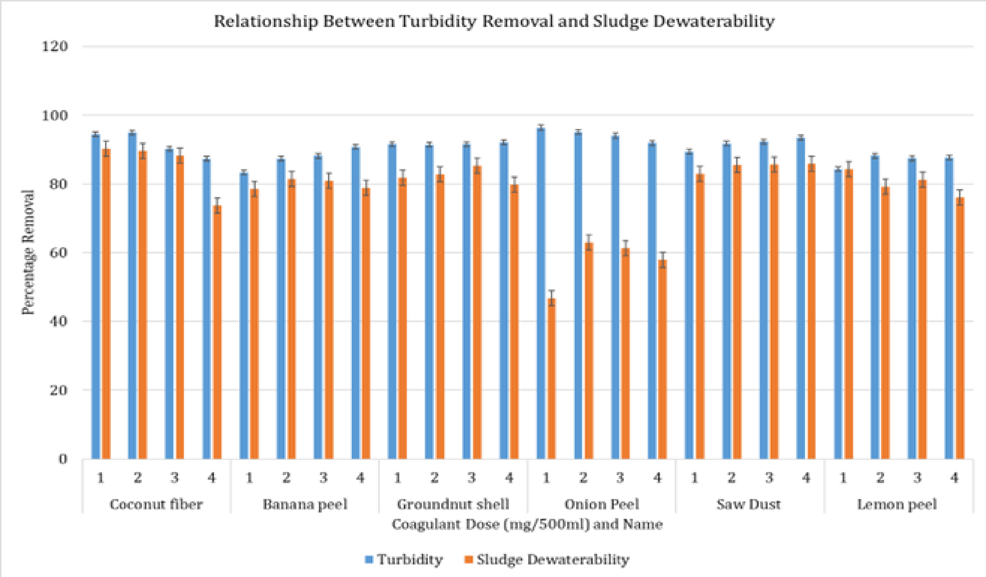

Fig. 5. *Comparison between turbidity removal and sludge dewaterability*

Table 3. *Statistical Analysis ANOVA of turbidity removal and sludge dewaterability*

S.No	Source of Variation	DF	SS	MS	F Value	Prob>F
1	Comparison of turbidity removal by 6 coagulants*	5	167.821	33.56	6.247	0.0015
	Error	18	96.705	5.372		
	Total	23	264.526			
2	Comparison of Sludge Dewaterability by 6 Coagulants*	5	2245.799	449.159	19.640	<0.0001
	Error	18	411.641	22.868		
	Total	23	2657.441			

DF = Degrees of Freedom; SS = Sum of Squares; MS = Mean Square* banana peel, onion peel, coconut fiber, groundnut shell, saw dust, lemon peel

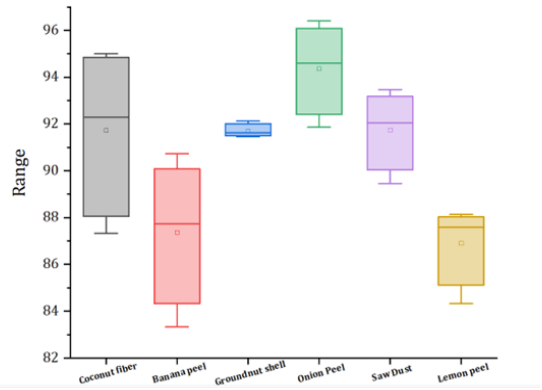

Fig. 6. *Origin plot of ANOVA for turbidity removal of synthetic turbid water by 6 coagulants*

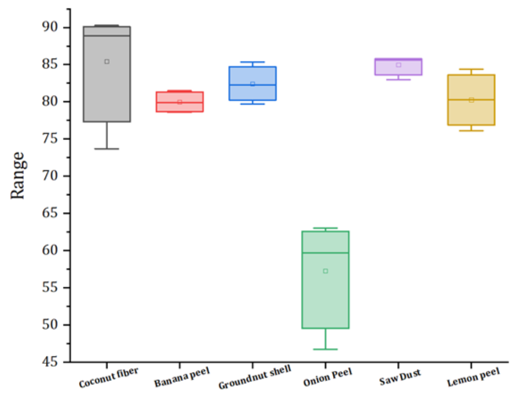

Fig. 7. *Origin plot of ANOVA for Sludge Dewaterability of synthetic turbid water by six coagulants*

Though the compact clumps of higher age are quite strong and are fast in settling, the presence of smooth surfaces and difficulty in forming larger aggregates in mild hydrodynamic conditions in the settling tank owing to the lack of irregular materials on their surfaces make them less effective to sweep smaller particles (Eriksson *et al*., 1992). From the study, it is observed that natural coagulants require a lower dosage to function, confirming their viability, which is in line with other studies (Ugonabo *et al*., 2024).

Table 3. presents the results obtained from the statistical analysis ANOVA (Analysis of Variance) over a comparison of turbidity removal by sixcoagulants (Fig. 6) and comparison of sludge dewaterability by sixcoagu-

lants (Fig. 7). Both the comparisons have showed a significant effect of coagulant dose on turbidity removal ($p<0.0015$) (Fig. 6) and also the effect of coagulant dose on the sludge formation ($p<0.0001$) (Fig. 7). These results indicated that the type of coagulant and dose significantly influenced turbidity removal and sludge produced.

Conclusion

Screening natural coagulants allow for the removal of ineffective coagulants that leave traces or alter the physicochemical properties of the treated water. The order of effectiveness in removing turbidity from the tested coagulants was as follows: Onion peel > coconut fibre > sawdust > groundnut shell > lemon peel > banana peel. The statistical analysis, using ANOVA to examine the correlation between the type of coagulant and turbidity removal, revealed a significant p-value (0.0015). The effectiveness of the tested coagulants in dewatering sludge was ranked as follows: coconut fibre > sawdust. The relationship between the type of coagulant and the ability of sludge to be dehydrated was analysed statistically using ANOVA and found to have a significant p-value of < 0.0001. Therefore, it can be deduced that the current screening method is an efficient approach that will result in saving a considerable amount of time and money in future research. Developing and implementing a strategy will facilitate the screening of naturally occurring coagulants for their use in water and wastewater treatment. The approach will save time and resources in selecting the coagulant and eliminating those that do not fit the criteria.

Conflict of interest

The authors declare that they have no conflict of interest.

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