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Research Article

Experimental screening and selection criteria of natural coagulants towards wastewater treatment

Swathi Dash
Department of Environmental Science, GITAM School of Science, GITAM (Deemed to be University), Visakhapatnam-530045 (Andhra Pradesh), India
Aman Raj
Department of Environmental Science, GITAM School of Science, GITAM (Deemed to be University), Visakhapatnam-530045 (Andhra Pradesh), India
Saritha Vara*
Department of Environmental Science, GITAM School of Science, GITAM (Deemed to be University), Visakhapatnam-530045 (Andhra Pradesh), India

*Corresponding author E-mail: vsjr08@gmail.com

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Abstract
The search for eco-friendly materials has greatly evolved ahead of basic requirements encompassing sustainable practices. The selection of such sustainable material requires procedural systematic screening tests to facilitate decision-making. Water and wastewater treatment processes involve several chemicals, and they need to transition from commercial to natural materials owing to their environmental and economic concerns. The present study aimed to select and screen natural coagulants for wastewater treatment. The criteria assessment factors for the present study were easy availability, economic value, turbidity removal efficiency, and reduced sludge generation with high dewaterability. A standard jar test apparatus was used for the coagulation experimental runs. The physicochemical parameters were analyzed using standard methods. The results presented positive insights into the efficiency of tested natural coagulants, with the least turbidity removal of 83.3% by rice husk at 1gm/500ml and the highest being 96.4% by onion peel at 1gm/500ml. The sludge obtained after treatment with natural coagulants has presented an excellent dewaterability, with the least being 29.17% by tamarind seeds at 1gm/500ml and the maximum being 90.2% by coconut fibre at 1gm/500ml. From the study, was concluded that the screening method promoted the selection of the best coagulant type and dose and reduced the efforts and time needed to eliminate the non-performers.

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

INTRODUCTION

Exploring novel materials will open fresh avenues for new goods such as natural coagulants, innovation, and the evolutionary progression of current materials to deliver enhanced and exceptional economic performance. The first step of the process would be to start with a comprehensive list of materials available, followed by selection; failing to do so might be a lost opportunity. Generally, acceptability will usually be for more than one material, and the final selection will be a compromise that includes pros and cons. The vast number of materials combined with the complexity of inter-relationships among various selection parameters make selecting materials challenging (Ashby, 1994; Nellippalli *et al*., 2020).

Some conventional methods for material selection were proposed by Sapuan and Abdalla, 1998, who used a knowledge-based expert system for selecting polymeric-based composite materials for a pedal box system (Shaharuzaman *et al*., 2021). In 2007, Kumar and Singh used a knowledge-based expert system to select materials for progressive dye components (Sinwar *et al*., 2023). Findik and Turan (2012) have developed a weighted property index method for selecting materials for designing lighter wagons. Their method has chosen five properties as necessary parameters: cost, specific stiffness, density, wear, and corrosion resistance, and it was improved using the weighted property index method (Sharma *et al*., 2022). Further, Merayo *et al*. (2019) investigated strategies of artificial intelligence for material selection in manufacturing processes.

Selection of material generally involves exploring the best combination of process attribute profiles and material along with the efficiency required by the material for a designed task. The crucial process in material selection is establishing a correlation between the function and material. Ashby (1994) presented the following strategy for selecting materials: Translation, screening, ranking, and documentation (Deng and Yu, 2022).

The translation stage involves investigating objectives, constraints, free variables and functions, which, when combined, will yield an appropriate material for selection. Screening essentially involves eliminating materials that do not fit owing to some attributes falling outside the constraints. Ranking is identifying the screened materials that are best suitable for the task. The criterion for ranking is the material's ability to do successfully in a particular application. Documentation essentially involves details of only those few materials that have successfully cleared the previous steps, which can be sought to enhance the work's feasibility (Ahmed *et al*., 2023).

The intrinsic relationship between ecosystem stability and biodiversity is a fundamental concept of ecological research. Availability of diverse materials for a defined purpose offers ecological stability and sustainability, avoiding the exploitation of selected materials or resources, thus mitigating environmental impacts, promoting resource efficiency, and contributing to the overall resilience of ecosystems and human systems (Craven *et al*., 2018). Hence, research on materials that can prove to be good natural coagulants is a continuing process. Screening these coagulants aligns with the sustainability goals as it involves resource planning, management and conservation, which address the sustainability challenges in an integrated manner. Hence, screening for natural coagulants will benefit in terms of effective use of resources, enhanced efficiency, reduced waste and increased value (Alam Bhuiyan and Hammad, 2023).

Throughout history, natural coagulants have played a significant role in water treatment. Their efficiency is governed by criteria such as i) Dose of the coagulant used, ii) Type of the coagulant, iii) Preparation and use of the coagulant and iv) Quality of the raw water.

The process of using plants as sources of natural coagulants includes i) identifying or screening the plants having coagulating properties; 2) extracting coagulant protein; 3) optimizing the coagulant for a particular source of water or wastewater (Ahmad *et al*., 2022; Saleem and Bachmann, 2019). Natural coagulants for water treatment practically date back to the existence of human civilization. The use of contemporary coagulants has progressively developed over the years through scientific exertions. However, the foundation of their development remains embedded in conventional or historic practices. Nevertheless, ancient wisdom has been the basis for the contemporary coagulation process and will remain an important source in the future (Charcosset, 2022; Said *et al*., 2023; Duggireddy and Pisharody, 2024).

Undeniably, research on natural coagulants has been experiencing resurgence in interest and attention in recent times, which reflects the recognition of the untapped potential of natural coagulants in addressing complex challenges of water and wastewater treatment (Simon and Joshi, 2021; Saleh *et al*., 2022; Knap-Bałdyga and Żubrowska-Sudoł, 2023). The present study aimed to select and screen for natural coagulants, and the criteria assessment factors considered for the present study were easy availability, economic value, turbidity removal efficiency, and reduced sludge generation with high dewaterability.

MATERIALS AND METHODS

Natural coagulants were collected from the source of generation. A total of six coagulants were selected for the present study: Rice husk, Saw dust, Lemon peel, Tamarind seeds, Coconut fiber and Onion peel. Rice Husk, tamarind seeds and Coconut fibre were collected from the rural areas around Visakhapatnam District, while sawdust was collected from a wooden furniture workshop on the outskirts of the city, the lemon peels and onion peels were collected from restaurants in the city. Analytical-reagent-grade chemicals were used to analyze water samples pre- and post-coagulation and prepare turbid water (Kaolin powder).

Preparation of coagulants

The coagulants were washed with tap water, oven-dried, and ground to a fine powder using the grinder. The powders thus obtained were stored in air-tight bottles at room temperature for further use.

Preparation of synthetic turbid water

Stock solution of kaolin was used for the preparation of synthetic turbid water by adding 10g of Kaolin to one litre of distilled water and was mixed by rapid mechanical agitation of 100rpm for 30min to obtain a uniform suspension of kaolin. Further, for complete hydration, the suspension was kept under quiescent conditions for 24 hours (Zainol *et al*., 2022). Various volumes were taken from the stock solution and dissolved in distilled water to prepare the desired turbid solution. In this study, 10 ml of the stock solution was dissolved in 500 ml of water to get 150 NTU solution.

After the selection of natural coagulants, screening of natural coagulants was done to treat physico-chemical parameters like Turbidity, pH, Conductivity, Colour, TDS and sludge with a set of coagulant doses. The present study focuses on screening of innate natural coagulants for their efficiency in removing turbidity and generating sludge. Six coagulants available abundantly disposed of as waste were selected for the study. A Jar Test Apparatus was used to test the efficiency of coagulation. From the tested coagulants, the ones that present the best efficiency were adopted for further studies. Hence, the present study considered the process for screening coagulants with effective removal capabilities. The efficacy of coagulants in removing turbidity and the amount of sludge generated was evaluated.

Water analysis

The following water quality parameters pre- and post-coagulation were considered for the study and analyzed below. All the analysis were conducted per the standard APHA methods, (2017) (Table 1).

Coagulation experiments

A conventional Jar test apparatus was used to test the efficacy of the coagulants under study. The 4 beakers were filled with 500ml of kaolin suspension each. To these 4 doses of each coagulant i.e. 1gm, 2gm, 3gm and 4gm, were added, i.e. a total of 6 sets of experimental runs in batch mode were carried out in duplicates. The samples were then made to be rapidly mixed initially at 80 rpm for 2 min, followed by 30 rpm for 30 min slow mixing. Then, the samples were kept under quiescent conditions for 30 min. Filtered samples were taken for post-coagulant analysis. The turbidity reduction percentage was calculated using the following equation (Saritha *et al.*, 2019).

$$Turbidity\ Reduction\ (\%) = \frac{Initial\ Turbidity - Final\ Turbidity}{Initial\ Turbidity}\ X\ 100$$

Eq.1

Wet and dry sludge

The weight of sludge obtained from the filtering water sample post-coagulation was considered as wet sludge. This wet sludge was oven-dried for 6 to 8 hours at 150^0C to remove the moisture content, and the resultant was considered dry sludge.

Statistical analysis

The influence of coagulant concentration (ppm) on turbidity removal percentage and sludge formed was evaluated using Analysis of Variance (ANOVA).

RESULTS AND DISCUSSION

The present study focuses on screening innate natural coagulants for their efficiency in removing turbidity and generating sludge. Fig. 1 represents the pH screening of selected natural coagulants; a decrease in pH changes was obtained with lemon peel with all 4 sets of doses, followed by sawdust.

All the coagulants tested brought changes in pH after the treatment. The rice husk and sawdust recorded a meagre increase of 0.1 to 0.3. A significant change was brought by lemon peels (reduction from 1 to 3 units), tamarind seeds and onion peels (increase from 1.0 to 1.8 units). Lemon peels contain abundant citric acid (Torrado *et al*., 2011). The reduction in pH is credited to citric acid, a tricarboxylic organic acid found in lemon peel (Li *et al*., 2024). The increase in pH by tamarind seed powder can be due to the presence of –OH function groups (Zainol *et al*., 2021). Slight changes in pH were reported by previous studies, where a decrease from 7.5 to 7.3 was noted (Abood *et al*., 2017; Kingue *et al*., 2023).

Fig 2 presents conductivity removal obtained after treatment with natural coagulants, the inclusion of conductivity was seen after treatment and in a few doses (1,2g of rice husk, 2,3g of lemon peel and all doses of sawdust), there was a totally no change in conductivity. Among the tested coagulants, saw dust did not alter the conductivity, which might be attributed to the ineffective destabilization and hydrolysis (Ntwampe, 2021). At lower concentrations of 1 and 2 gm/L rice husk and lemon peel, conductivity did not change. However, as the coagulant dose increased, both coagulants induced conductivity (- 16.6%). Coconut fibres induced or enhanced conductivity to the highest level (-66.6%), followed by tamarind seeds and onion peel (-50%).

Electrical conductivity presents the amount of substances dissolved in the solution. The increase in conductivity could be due to dissolved ions in the water, enabling the conduct of electricity. Ogunshina *et al*. (2023) presented that the conductivity has enhanced in the treated water (401.07 to 1000mS/cm) using *Moringa oleifera* as the coagulant. Further, Putra *et al*. (2020) presented that moringa protein converts to positive molecules upon dissolving at pH 7-8. Protein is a zwitter ion and polyelectrolyte compound composed of carboxylic acid and an amine group; it will form positive ions upon dissolving in water. The release of H+ ions enhanced the treated water's conductivity.

Fig 3 presents turbidity removal, total dissolved solids, and colour removal by treating them with natural coagulants. Highest turbidity removal was obtained with onion peel at 96.4%, 95.13%, 94.06%, and 91.86% and the lowest was seen in rice husk with 83.33%, respectively. It was noted that the coagulants induced TDS and have shown a meagre change in the colour after treatment. The highest turbidity removal (96.4% at

Table 1. Methods of physico-chemical analysis

Parameter	Method of Analysis
Colour	IS 3025, part 4
Turbidity	APHA 2130 B
pH	APHA 4500 H+ B
Electrical Conductivity	APHA 2510
Total Dissolved Solids	APHA Standard Method 2540C
Sludge weight	APHA Standard Method 2710

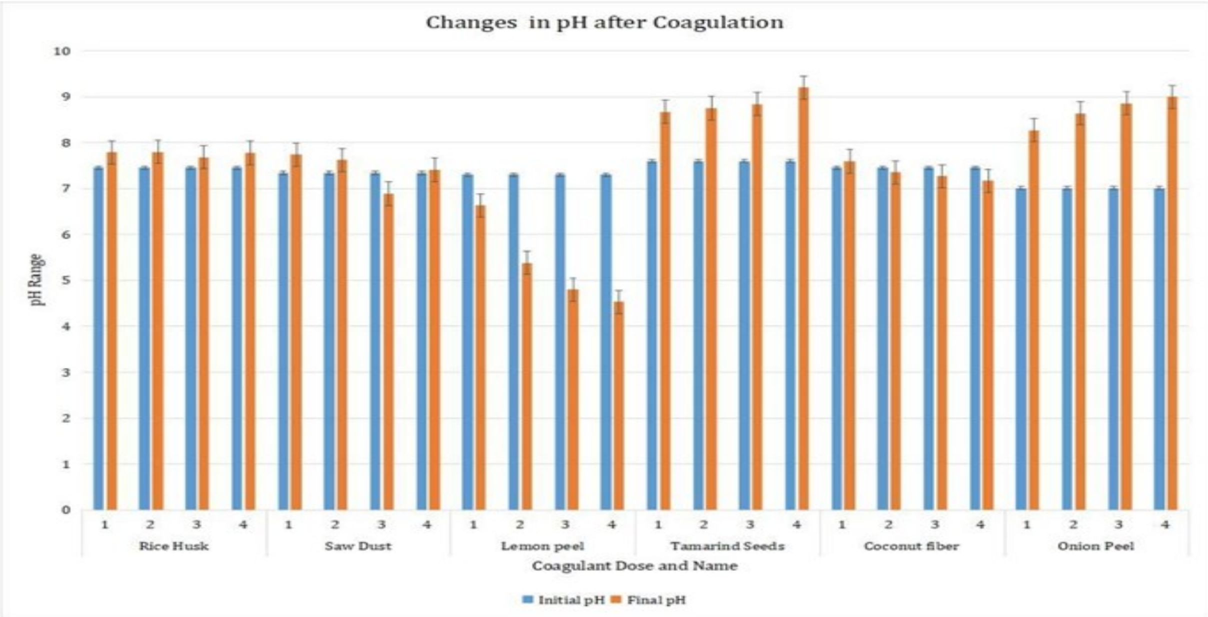

Fig. 1. *Changes in pH after Treatment with Coagulants*

1g/L) was recorded by onion peel, which decreased with the increase in coagulant dose. Similar trend was observed with coconut fiber and tamarind seeds (94.4% at 1g/L). The results are in agreement with studies by Febrianti *et al*., 2024, where they found that the turbidity removal was inversely proportional to coagulant dose since larger doses of coagulant (Aloe vera) can induce the formation of agglomerates that are not optimum and hence lesser removal of turbidity (Mujariah *et al*., 2017).

In contrast, the turbidity removal efficiencies increased with increasing coagulant dose for coagulants rice husk, sawdust and lemon peel, with sawdust recording the highest removal (93.46% at 1g/L). This is attributed to coagulant dose being directly proportional to the cation exchange capabilities of these coagulants, destabilising the solution's colloids (Karnena and Saritha, 2022). Noteworthy removal of turbidity from raw water by innate coagulants like *Cicer arietinum, Dolichos lablab* and *Moringa oleifera* was previously reported (Jasim *et al*., 2022). Mohd-Asharuddin *et al*. (2017) illustrated that the most significant turbidity removal (29%) was obtained with 200 mg/L of cassava peel starch. Upon increasing the dosage, they observed an increase in turbidity removal. This can be caused due to the complex coulombic attraction among the colloidal particles and the coagulant, which repel negatively charged particles. Moreover, lower concentrations of positively charged ions of the coagulant probably are not enough for neutralizing the surfaces of colloids

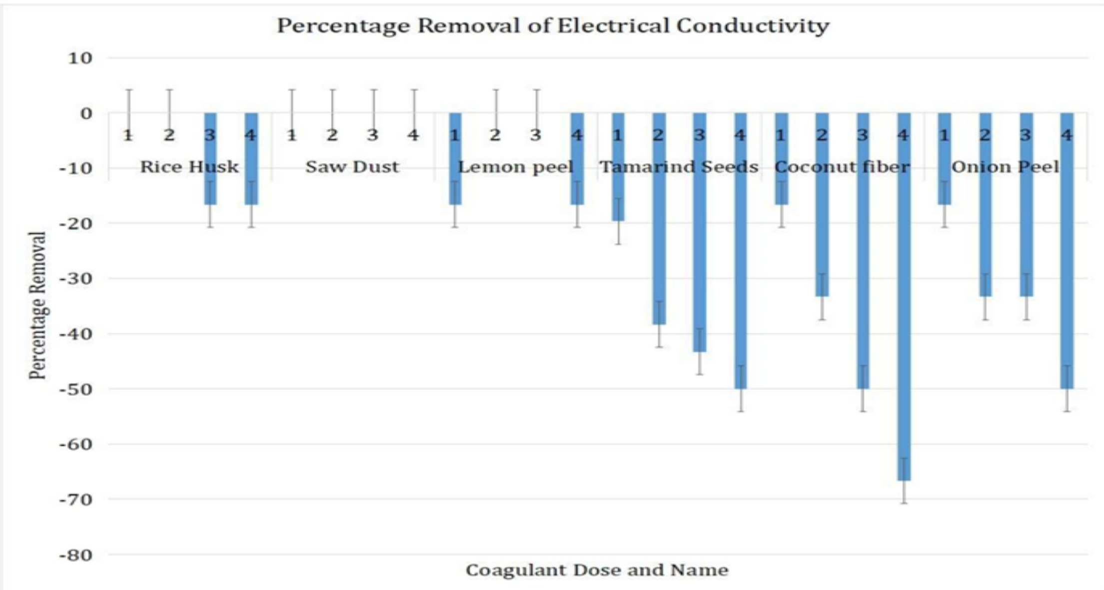

Fig. 2. *Percentage removal of Electrical Conductivity by Coagulants*

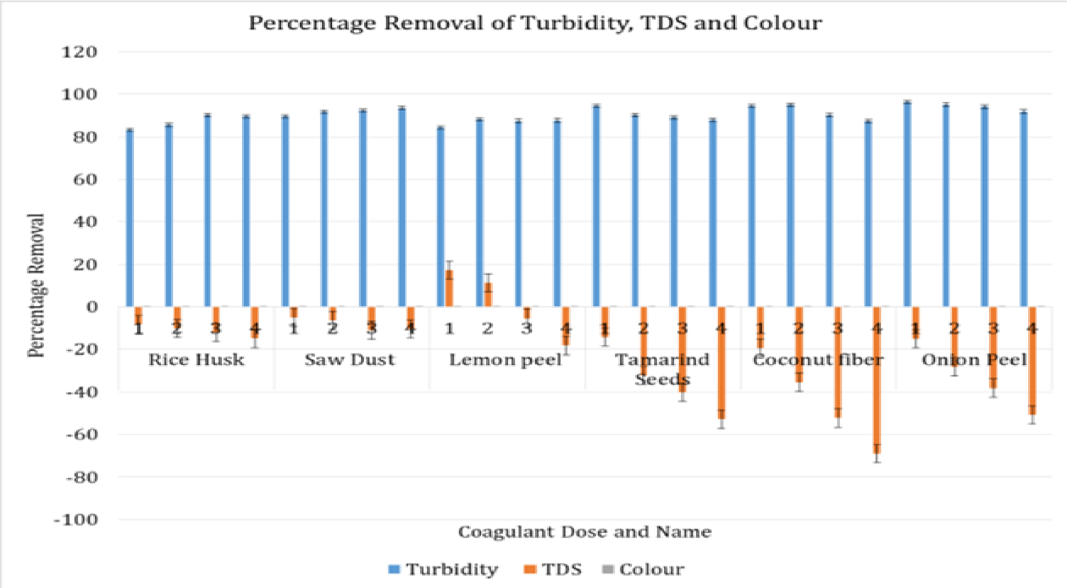

Fig. 3 *Percentage removal of turbidity, total dissolved solids and colour*

(Mohd-Salleh *et al*., 2020). The results were in line with the previous studies, where 91% reduction in turbidity was obtained with blended coagulant rockmelon seeds (Joaquin *et al*., 2024).

Except for lemon peels which showed reduction in TDS at lower doses of 17.31% at 1g/L and 11.31% at 2 g/L. All the other tested coagulants have induced total dissolved solids (TDS) in the treated water. Coconut fiber induced the highest TDS (-69.15%), followed by tamarind seeds (-52.9%) and onion peels (-50.9%). Least TDS (-5.04%) was induced by saw dust at 1g/L. Coconut coir, when used as a biocomposite for removal of various physic-chemical properties resulted in 16% removal of TDS and they have also reported that the removal rates increased with an increase in the dose of the adsorbent (Gutub *et al*., 2013). The studies conducted by Febrianti *et al*. (2024) showed an increase in TDS when the coagulant was extracted using water. They attributed the increase in TDS value to excessive coagulant doses of 1 and 1.5ml, which caused the reformation of colloidal particles.

Colour removal by all the tested coagulants was very meagre (0.01 to 0.07%), with lemon peels, tamarind seeds and coconut fibre highest at 4g/L. Activated carbon from tamarind seeds increased dye adsorption up to 1.0gm but showed no significant changes after 1gm, showing equilibrium. This is attributed to the enhanced surface area and number of adsorption sites. Adsorption of methylene blue using iron oxide nanoparticles and onion peels resulted in 97% removal within 30 minutes (Abid *et al*., 2021). Nguyet *et al*. (2020) presented that purified coconut fibre possessed a decent adsorption property at optimal conditions of 0.7g dose, 8 pH, 30^{0}C temperature and a contact time of 24h (Nguyet *et al*., 2020). Activated carbon from lemon peels showed an increase in the deduction of methylene blue dye with doses from 0.01g to 0.1g (Ramutshatsha-Makhwedzha *et al*., 2022).

Table 2 presents the results from the variance analysis over the type of coagulant and the tested parameters, finding that the tested coagulants have significantly removed turbidity and have also shown a noteworthy effect on sludge formation ($p<0.05$). The results obtained are in accordance with the studies of Villabona-Ortiz *et al*., 2023 (Fig. 4 and 5).

Fig. 6 represents the weights of sludge generated after treatment with the natural coagulants. Maximum amount of sludge generated was with coconut fiber, around 32.55g followed by onion peel with 28.97g and minimum sludge was generated with rice husk of 4.13g, respectively. Fig. 7 represents the turbidity removal and sludge dewaterability obtained by treating with natural coagulants, reduction in turbidity removal was observed as the dose of coagulant enhanced for tamarind seed, coconut fiber and onion peel; an increase in turbidity removal obtained in rice husk, sawdust and lemon peel as the dose of coagulant increased. An increase in sludge dewaterability was reported as the dose of coagulant was enhanced. The highest turbidity removal was 96.4%, the lowest was 83.33%, minimum removal of sludge dewaterability was obtained at 29.17%, respectively.

Sludge generated by natural coagulants

Water and wastewater treatment using coagulation and flocculation results in the generation of sludge, which consists of residual solids containing coagulants and contaminants. Due to its environmental impact, it is crucial to manage and dispose of the sludge properly (Badawi and Zaher, 2021). Sludge dewatering is the process of reducing the moisture content of the sludge, thus dropping its volume for further treatment or dis-

Table 2. Statistical analysis using ANOVA

S.No.	Source of Variation	DF	SS	MS	F Value	Prob>F
1	Comparison of Turbidity Removal by 6 Coagulants	5	167.73	33.54	4.855	0.0055
	Error	18	124.35	6.908		
	Total	23	292.08			
2	Comparison of Sludge Dewaterability by 6 Coagulants	5	2187.72	437.54	10.54	<0.0001
	Error	18	746.995	41.499		
	Total	23	2934.72			

DF (Degrees of freedom in the source); SS (Sum of squares due to the source) MS (Mean sum of squares due to the source); F (F-statistic).

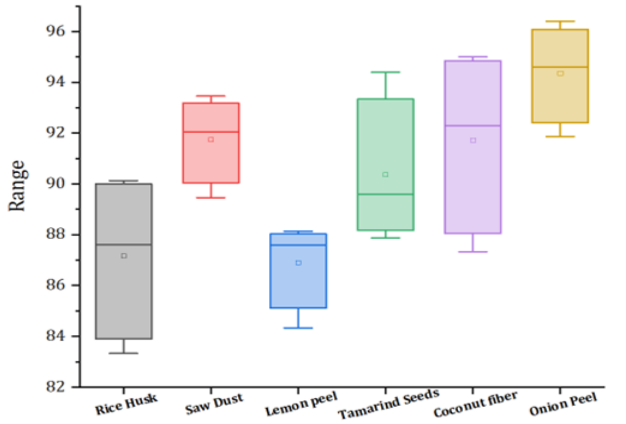

Fig. 4. *Origin plot of ANOVA for turbidity removal by 6 coagulants*

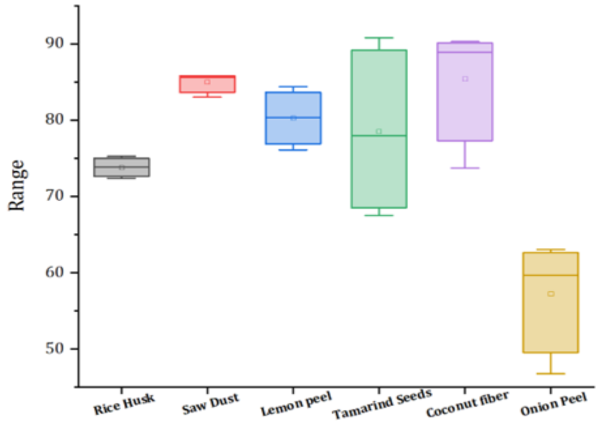

Fig. 5. *Origin plot of ANOVA for sludge dewaterability by 6 coagulants*

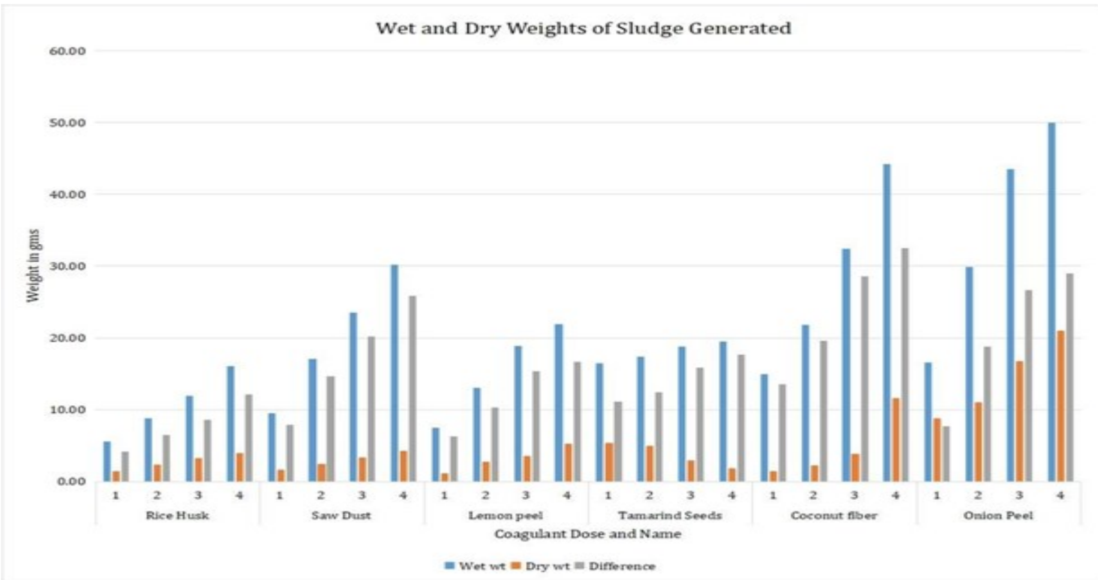

Fig. 6. *Wet and dry weights of the sludge generated*

carding (Wei *et al*., 2018). The advantage of using organic polymers is that they not only produce less sludge but also the produced sludge has better dewatering characteristics (Dayarathne *et al*., 2021). In the present study, all the tested coagulants have not only produced lesser sludge, but the dewaterability of the sludge after moisture removal ranged from a minimum of 29.7% (Tamarind seeds 1g) to 90.27% (Coconut fibre 1g). Further, being organic, the sludge generated from natural coagulants can be used as a soil conditioner. The sludge nutrient content, organic matter and its binding properties can enhance soil quality, promoting plant growth if applied in suitable quantities, following controlling guidelines (Zhou *et al*., 2021).

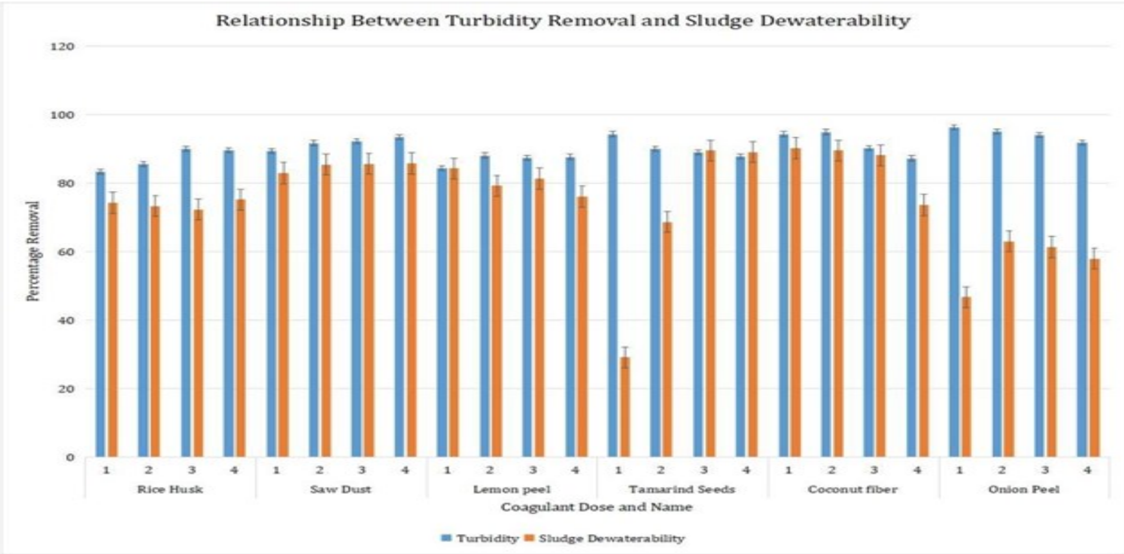

Fig. 7. *Comparison between turbidity removal and sludge dewaterability*

The order of average sludge dewaterability by individual coagulants was Coconut fibre > Saw dust > Lemon peel > Rice husk > Tamarind seeds > Onion peel.

Scope of plant-based coagulants in the treatment of water

Plant-based coagulants are a primary and principal source of water treatment for the significant number of people across the world (Owodunni and Ismail, 2021; Koul *et al*., 2022). In the highly competitive environment of contemporary water research, natural coagulants provide unique functionality indispensable for water and wastewater (Al-Jadabi *et al*., 2023; Sahu *et al*., 2023).
Scientifically, it is known that natural coagulants possess a higher number of polymers than chemical polymers. The polymers from natural coagulants have a high molecular weight, which can form larger, complex structures resulting in larger flocs that aid in the exclusion of more colloids, suspended particles and other impurities, enhancing the efficiency of water and wastewater treatment (Tijjani Usman *et al*., 2023). Additionally, coagulants from plant sources contain a diverse array of functional groups that interact with contaminants in water through varied mechanisms contributing to their coagulation (El-taweel *et al*., 2023; Jagaba *et al*., 2023; Hu *et al*., 2024).
Despite tremendous progress in water treatment by coagulation and coagulants, many treatment units and researchers are expanding their screening programmes for natural coagulants to ascertain active coagulating agents (Diver *et al*., 2023; Muniraj *et al*., 2023). Identifying coagulating activity from plants/their parts is a long process, including selecting plant, screening for coagulant protein, experimental evaluation of coagulation efficiency and optimization (Louhichi *et al*., 2024; Benalia *et al*., 2024). Given the promising results and efficiency shown by coagulants previously studied, the research for evaluation of plant source coagulants will see a hike in the future (Knap-Bałdyga *et al*., 2023; Ragio *et al*., 2023; Hadadi *et al*., 2023).

Conclusion

Turbidity removal, amount of sludge generated, and dewaterability were the parameters for screening natural coagulants. This process of selecting natural coagulants enabled the elimination of coagulants which were ineffective in turbidity removal and also resulted in greater sludge or leaving traces in treated water, resulting in inducing or changing physicochemical parameters such as pH, electrical conductivity, colour, total dissolved solids and turbidity. The efficiency of the tested coagulants Rice husk, Saw dust, Lemon peel, Tamarind seeds, Coconut fiber and Onion peel varied owing to the type of coagulant. However, the efficiency in terms of turbidity removal was in the order of onion peel > coconut fibre > tamarind seeds > saw dust > lemon peel > rice husk. On the other hand, the efficiency of the tested coagulants in generating good dewaterable sludge was in the order of coconut fibre > lemon peel> saw dust> rice Chunk> tamarind seeds> onion peel. Thus, the present screening approach was considered an effective method that will save significant time and cost in further studies with reference to the selection of appropriate coagulants for treating a specific type of wastewater.

Conflict of interest
The authors declare that they have no conflict of interest.

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