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Hybrid system using local plant as sustainable coagulation-flocculation process

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ABSTRACT

Coagulation-flocculation is widely used in the treatment of water and wastewater to remove suspended and colloidal particles. Most chemical coagulants used are synthetic and after treatment, the sludge residuals will be toxic and unable to recovery as fertilizer. This study used natural coagulants which are safe because after treatment, the sludge residual no toxic. Seven types of natural coagulants were selected from Iraqi plants to investigate the 500 NTU turbidity removal prepared from Kaolin powder. The experiment parameters were coagulant concentrations ranging from 0 to 10.000 mg/L, fast mixing (200 rpm) for 5 minutes, slow mixing (30 rpm) for 15 minutes, and 25 minutes of sedimentation time. Thereafter, a hybrid system is investigated for the best three natural coagulants with aid alum concentrations of 5 mg/L and 10 mg/L. Results show that the three most effective natural coagulants had turbidity removal of 48, 47, and 47 for palm fiber, palm pith, and watermelon rinds, respectively, at 500 mg/L concentration. For a hybrid system, the best one is with 10 mg/L alum aid with 66% turbidity removal efficiencies for watermelon rinds at 500 mg/L concentration. Natural coagulant is a sustainable solution for a safe environment.

Keywords: local plant, natural coagulant, turbid water, sustainable solution, safe.

INTRODUCTION

The problem of water shortage can be control when save usage, decrease pollution, and treatment of contaminated water before discharge into the environment. The simple way for water treatment is coagulation, particularly by bring the suspended particles together to be in flocs and removed thereafter by sedimentation (Kalibbala et al., 2023). The most frequently used chemical for coagulation in water treatment plants is alum. During wastewater treatment, the addition of chemical coagulants produces non-biodegradable sludge, thereby causing health and environmental hazards (Hounsinou et al., 2023). Natural coagulants are safe for the public and ecosystem because they are organic (i.e., easy to degrade), affordable, local, and environmentally sustainable.

The use of natural coagulants (low molecular weight) with chemical coagulants (high molecular weight) enhances aggregation configuration, thereby providing good stability for the coagulation–flocculation process (Ang and Mohammad, 2020). Water and wastewater treatment plants are constantly searching for sustainable technology of integer by utilizing natural materials to achieve high-quality treatment with minimum residual toxic sludge. To obtain high performance in coagulation-flocculation process, natural coagulants with addition chemical coagulant in hybrid treatment process was adopted (Mutar et al., 2022). Hybrid treatment can be implemented with the coagulation-flocculation process by natural coagulants with inorganic coagulant, such as alum, to increase natural coagulants efficiency and decreased residual toxic sludge (Ang and Mohammad, 2020). Shak and Wu (2015) examined the combination of alum (1.15 g/L) with *Cassia obtusifolia* seeds (2.47 g/L) in the treatment of palm oil mill effluent and reached to remove of total suspended solids and chemical oxygen demand removals up to 81.58% and 48.22%, respectively (Shak and Wu, 2015). The use of green coagulation–flocculation process in water and wastewater treatments can be the potential for recovery and recycling of primary treatment sludge to use as fertilizer. Recovery and recycling are sustainable options for water treatment based on sustainable development goal (SDG) 6 for safe drinking water. Suspended particles and colloids in water and wastewater are negatively charged, whereas natural coagulants from seeds, leaves, animals, bacteria, fungi, and algae are positively charged, thereby enhancing floc aggregation by active components, such as polysaccharides, protein polymers, and certain functional groups, such as hydroxyl and carboxyl groups (Husen et al., 2024). Considerable research on the green coagulation–flocculation process has been conducted related to water and wastewater treatment, as summarized in Table 1. Desta and Bote (2021) found that 0.1 g of *Moringa oleifera seeds* can be result in 98%, 90.76%, and 65.8% removal efficiency for turbidity, color, and chemical oxygen demand respectively, from wastewater (Desta and Bote, 2021). For a hybrid system, Asharuddin et al. (2023) used alum and tapioca peel starch (TPS) to reach the highest turbidity removal of 90.32% by mixing 7.50 mg/L alum with 50 mg/L TPS (Asharuddin et al., 2023). Table 1 presents the studies conducted on natural coagulant application. To the best of our knowledge, the combined use of palm fiber, palm pith, and watermelon rinds with alum is not investigated yet. Thus, hybrid system was adopted to investigate the performance of natural coagulant with Alum which is effective on water treatment. This study aims to investigate the best local plants using the coagulation–flocculation process for turbidity removal and utilizes a hybrid system that can be used to improve the efficiency of natural coagulant with the aid of alum. Natural coagulant is an interesting unconventional method to developing countries like Iraq for aquaculture industry because it is economy, eco-friendly, and recovery of residual sludge as fertilizer.

MATERIALS AND METHODS

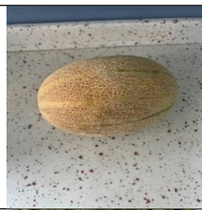
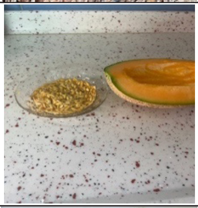
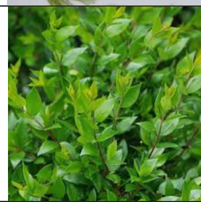
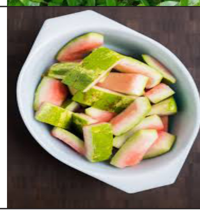
Natural coagulant selection

As shown in Table 2, seven plants were chosen as natural coagulant and different parts were

Table 1. Studies on natural coagulant application

Natural Coagulants	Treatment	Coagulant Dosages	Removal Efficiency	References
Moringa oleifera (seeds)	Palm oil mill effluent	2000 mg/L	TSS: 95.42 % Turbidity: 88.30%, Color: 90.15 % NH3-N: 89.81% Oil & grease: 87.05 %	(Jagaba et al., 2020)
Papaya (seeds)	Tulte River (392.7NTU)	20 mL	Turbidity: 96.19%.	(Yimer and Dame, 2021)
Leucaena leucocephala (seeds)	Turbid water (450 NTU)	120 mg/L	Turbidity: 93.05%	(Alnawajha et al., 2024)
Trigonella foenum-graecum *Abelmoschus esculentus*	Palm oil mill effluent	4.09 mg/L 57.69 mg/L	Turbidity: 94.97% TSS: 92.70 % COD: 63.11 %	(Lanan et al., 2021)
Cicer arietinum *Strychnos potatorum* *Moringa oleifera*	Dairy wastewater	400 mg/L 800 mg/L 400 mg/L	Turbidity: 74.23% BOD: 63.2%. Turbidity: 75.62% BOD: 72.1%. Turbidity: 65.6% BOD: 70.9%.	(Deepa et al., 2022)
Conocarpus lancifolius *Azadirachta indica* *Nerium oleander*	Turbid water (500 NTU)	5000 mg/L 5000 mg/L 1000 mg/L	Turbidity: 75.5% Turbidity: 67.2% Turbidity: 57.2%	(Khalid Salem et al., 2023)
Grape seed + *Austrocylindropuntia mucilage*	Turbidity: 250 NTU Congo red (CR) azo dye: 5 mg/L	0.45 mg/L + 6 mL/ L	Turbidity: 95.74% Congo red (CR) azo dye: 99.36 %	(El Gaayda et al., 2024)
- *Beta vulgaris L.* (stem) - *Alhagi graecorum* - *Prunus armeniaca*	Turbid water (500 NTU)	1000 mg/L 3000 mg/L 7000 mg/L	Turbidity: 56% Turbidity: 67% Turbidity: 62%	(Hatim et al., 2024)
Ipomoea batatas (leaves)	Turbid water (250 NTU)	10 g/L	Turbidity: 90%	(Kusuma et al., 2021)
Crescentia cujete (fruit shell)	Tano River. (13.28 NTU)	1 g/L	Turbidity: 83.4%	(Boakye et al., 2024)

Table 2. Selected the plants for coagulants

Scientific plant names	Family names	Common names	Full plant profiles	Parts of the plants
Eucalyptus bark	myrtle	gum trees		
Melon seeds	*Cucurbitaceae* (gourd)			
Ceratophyllum	*Ceratophyllaceae*	hornwort		
Myrtus communis	*Myrtaceae*	myrtle		
Watermelon rinds	*Cucurbitaceae*	*Citrullus lanatus*		
Phoenix dactylifera	*Arecaceae*	Palm fiber		
Phoenix dactylifera	*Arecaceae*	Palm pith		

taken: eucalyptus bark, melon seeds, *Ceratophyllum* leaves, *Myrtus communis* submergent plant, watermelon rinds, palm fiber, and palm pith.

Preparation of natural coagulant

After collecting the selected plant parts, they were cleaned with distilled water and placed in an oven at 60 °C for 24 h. Thereafter, dry coagulant was ground and sieved to a fine powder and stored in a closed container (Hatim et al., 2024). To prepare a stock of coagulants with 100.000 mg/L concentration, 10 g of plant powder was added to 100 mL of distilled water, and mixed thereafter for 30 min (Kusuma et al., 2021). Subsequently, the extracted natural coagulant was separated from water by passing it through a double layer nylon cloth. The coagulant was filtered through a 0.45 µm mm ZELPA Belgium paper and kept the extraction in a Duran bottle. Thereafter, it is ready to use.

Turbid water

Kaolin powder was used to prepare turbid water with 500 NTU concentration produced by mixing 1.5 g of kaolin with 3 L distilled water in a container. To obtain homogeneous turbid water with concentration of 500 ± 50 NTU, the liquid was stirred for 30 minutes (Ahmad et al., 2022).

Jar test setup

The test was conducted using jar test (Lovibond-Germeny) with six 1-L beakers and paddle flocculator, and the working volume was 500 mL (Zaki et al., 2024). The experiment was conducted with the seven selected Iraqi plants after preparing a stock solution for each plant with 100.000 mg/L through water extraction. Six concentrations of natural coagulant at 0 (as control), 500, 1.000, 3.000, 5000, and 10.000 mg/L were ran to distinguish two of the concentrations that can be efficient for the coagulation–flocculation process for the next step of the research. The test was implemented with 200 rpm for 5 minutes as rapid mixing, and 30 rpm for 15 minutes as slow mixing. Following the rapid and slow mixing options, the suspended material was allowed to settle for 30 minutes (Boakye et al., 2024). To measure the turbidity of each beaker, a sample of 10 mL was taken from the top and measured using a turbidity meter (Lovibond, Germany). Equation 1 was utilized to determine the efficiency of turbidity removal (Lanan et al., 2021). To reduce reading error and compute the average value, three replicates were taken from each beaker.

$$\text{Turbidity removal } (\%) = \frac{Ti - Tf}{Ti} \times 100 \qquad (1)$$

where: T_i and T_f (NTU) are the initial and final turbidity, respectively, of the water.

Preparation of the hybrid system test

A stock solution of 500 mg/L hydrated aluminum sulfate ($A_{12}(SO_4)_3 \cdot 18\ H_2O$) (Thomas Baker-India) was prepared using distilled water and diluted into 5 mg/L and 10 mg/L concentrations to use in the hybrid system test, which was adopted according to optimum dosage (25 mg/L) used in water treatment plants (Qasim et al., 2000; Asharuddin et al., 2023). The three best local plants will process using the hybrid system as set up (as shown in Figure 1) with 500 NTU turbid water.

Turbidity removal in hybrid system with statistical analysis

Data of the hybrid system were analyzed using the Statistical Package for the Social Sciences (SPSS version 21 software, Inc. Chicago, USA) (Sharuddin et al., 2024). In this test, 95% confidence interval ($\alpha = 0.05$) was adopted for SPSS, and the significance difference was set at $p < 0.05$ to compare between groups. Analysis of variance (ANOVA) was used to check the natural coagulant alone and with 5 mg/L and 10 mg/L alum aid in the hybrid system in turbidity removal efficiency. The analysis was performed with one-way ANOVA and selected Turkey HSD in the post-hoc test to find significant differences in the results (Qays et al., 2023).

RESULTS AND DISCUSSIONS

Plant role as sustainable coagulant

Various plant parts (e.g., leaves, seed, bark, or pith) were selected for turbidity removal from water because they are environment-friendly. For palm fiber, palm pith, and watermelon rinds,

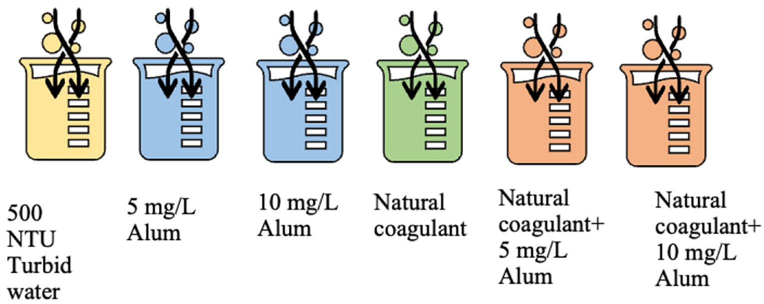


Figure 1. Hybrid system setup

removal efficiencies were 48%, 48%, and 47% compared with the control (only turbid water, 22 ± 2%), and palm fiber, palm pith, and watermelon rinds are the best comparison with other natural coagulants (Figure 2). Melon seeds, *Ceratophyllum*, and eucalyptus bark achieved 36%, 43%, and 43% removal efficiencies at 500 mg/L coagulant concentration. However, *Myrtus communis* obtained 42% removal efficiency at 1000 mg/L coagulant concentration compared with the control (only turbid water, 22 ± 2%) (Figure 3). Other studies, such as Fenugreek, used coagulant to treat palm oil mill effluent, and the results showed removal rates of 94.97%, 92.70%, and 63.11% in turbidity, TSS, and COD, respectively (Lanan et al., 2021). Table 3 shows the summary of the removal efficiencies if the seven selected plants with the coagulant concentration.

Hybrid sustainable system (alum plus natural coagulant)

To achieve high quality in water and wastewater treatment plants, the integration (hybrid system) of conventional and green technologies can be adopted in the coagulation–flocculation process. The coagulation performances of hybrid system are presented in Figure 4. The turbidity removal efficiencies of the three coagulants (i.e., palm fiber, palm pith, and watermelon rinds) used have been compared with 500 NTU turbid water and aided alum with 5 mg/L and 10 mg/L concentrations.

Accordingly, the treatment using natural coagulants alone showed undistinguished turbidity removal compared with chemical coagulants. Turbidity removal was observed higher when alum was added to the natural coagulant. The combined palm fiber, palm pith, and watermelon rinds with alum showed markedly performance owing to the integration of organic and inorganic coagulants led to maximum turbidity removal that reached 66%. The turbidity removal was 48, 52, 55% for palm fiber only, palm fiber + 5 mg/L alum, and palm fiber + 10 mg/L alum respectively. Regarding palm pith, the turbidity removal was 47, 57, and 60% for palm pith only, palm pith + 5 mg/L alum, and palm pith + 10 mg/L alum

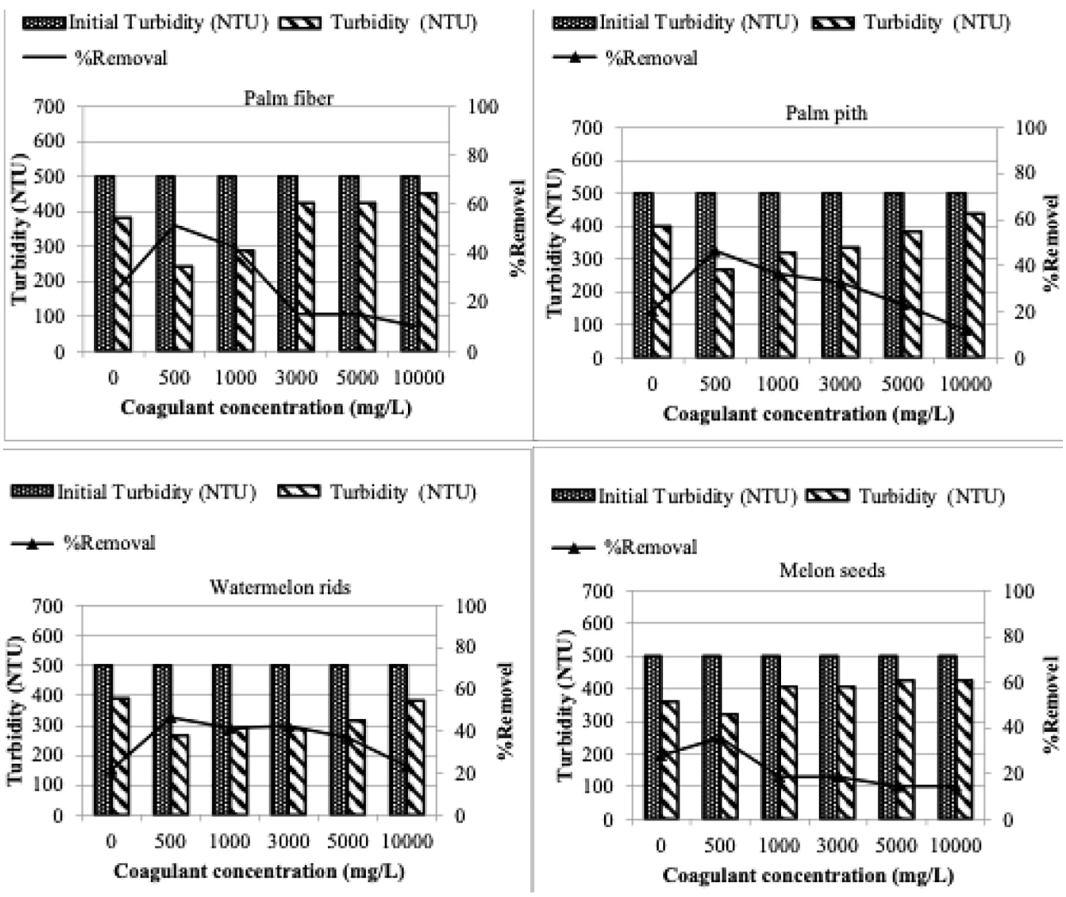


Figure 2. Turbidity removal for palm fiber, palm pith, watermelon rinds, and melon seeds

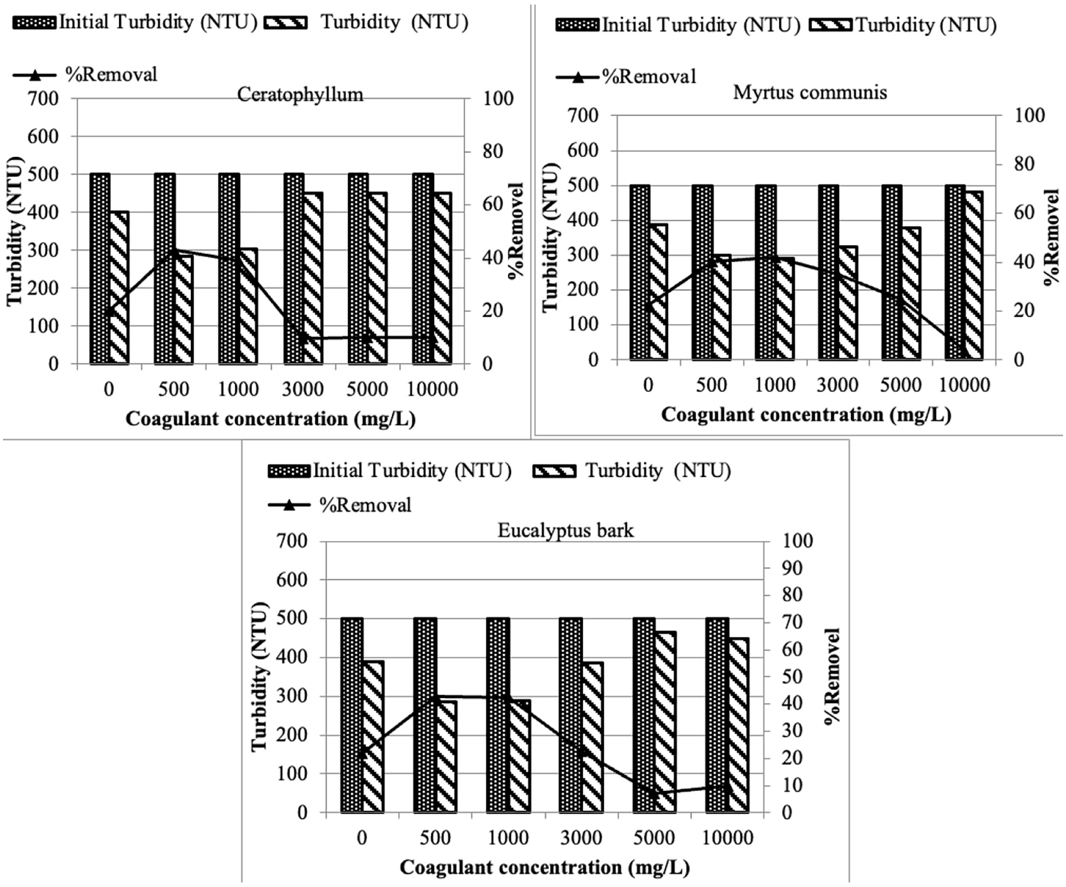


Figure 3. Turbidity removal for *Ceratophyllum, Myrtus communis*, and eucalyptus bar

Table 3. Best coagulant concentrations for the highest turbidity removal efficiencies

Natural plants	% Removal	Concentrations (mg/L)
Palm fiber	48	500
Palm pith	48	500
Watermelon rinds	47	500
Melon seeds	36	500
Ceratophyllum	43	500
Myrtus communis	42	1000
Eucalyptus bark	43	500

respectively. While turbidity removal was 47, 60, and 66% for watermelon rinds only, watermelon rinds + 5 mg/L alum, and watermelon rinds + 10 mg/L alum respectively.

For the three selected natural coagulants of palm fiber, palm pith, and watermelon rinds, there is significant different in 5 and 10 mg/L Alum aid compared to only natural coagulant ($p < 0.05$) according to ANOVA analysis. The high removal efficiency was in the hybrid system with 10 mg/L alum aid. Natural coagulant includes high amylose contents led to increase amount of amylopectin chains, which assist interaction between colloidal particles and starch that increases flocculation then removal efficiency (Asharuddin et al., 2023). In addition, natural coagulant include positive cationic elements which are enhanced the charge attraction with negative suspended particles for destabilization through the processes of adsorption and interparticle bridging.

The findings showed that the combination of alum in low concentration with natural coagulants is mostly suitable for high efficiency. This combination in the hybrid system was consistent with other studies' (Asharuddin et al., 2023) results, in which a decrease in inorganic coagulant (alum) within range (5–15 mg/L) was observed by combining organic natural coagulants with the coagulation–flocculation process. (Saqib et al., 2024) found that the hybrid system of alum + guava leaves coagulants (CAAGL) show a 96% coagulation efficiency in removing acid dyes from wastewater. The results showed the combine two coagulant species as hybrid system significantly affected the flocs size. This result proposed

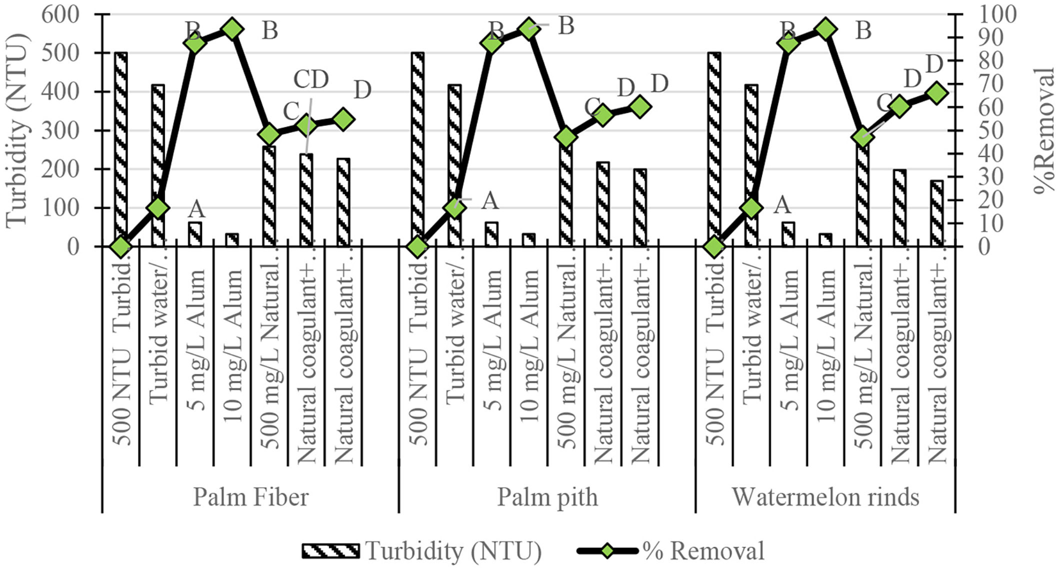


Figure 4. Variation of turbidity removal for palm fiber, palm pith, and watermelon rids alone and with aid alum. Differences in capital letters indicate significant differences at $p < 0.05$ in controlling turbid water, 5 mg/L and 10 mg/L alum, natural coagulant only, and hybrid system

that alum enhanced the particle collision and thus produced bigger aggregate size at lower Alum dosage of 5 and 10 mg/L.

CONCLUSIONS

The strategy of intergradation system to enhance the efficiency of the coagulation process (using the natural coagulant aid, alum) results in high turbidity removal. In this study, the addition of low alum concentrations with natural coagulant achieved high removal efficiency compared with only natural coagulant. The best coagulants tested were palm fiber, palm pith, and watermelon rinds, which reached 48%, 48%, and 47% turbidity removal efficiencies. When processed with the hybrid system, the best results for 500 mg/L watermelon rinds concentration plus 10 mg/L alum was 66% compared with 47% for watermelon rinds coagulant only. A hybrid system can be a sustainable solution to improve the efficiency of natural coagulants.

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