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EXPLORATION OF VARIOUS INDONESIAN INDIGENOUS PLANTS AS NATURAL COAGULANTS FOR SYNTHETIC TURBID WATER

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ABSTRACT

The availability of clean water is one of the biggest global problems, especially in developing countries. The simplest way to treat turbid water is by using coagulation. However, the utilization of chemical coagulants such as alum and ferrum has several drawbacks, including a high sludge volume and negative health impact when the water is consumed. Natural coagulants offer a better option, especially because of their availability, low price, lower sludge volume, and effectiveness comparable with chemical coagulants. In this study we utilize *Moringa oleifera*, *Carica papaya* and *Leucaena leucocephala* seeds, which are indigenous plants in Indonesia, as natural coagulants. An FTIR study was conducted to qualitatively identify the possible active coagulant agent in the seeds. The coagulant performance in removing turbidity of synthetic kaolin water was studied using jar test apparatus at various levels of coagulant dosage and pH. Functional groups of –OH, N-H, C=O, and primary, secondary and tertiary amides were identified in all the seeds. Neither the dosage nor the pH had an effect on turbidity removal when *M.oleifera* was used as the natural coagulant, but did have some effect on papaya and leucaena. The turbidity removal obtained in this study was comparable with other reported results, therefore it can be concluded that these seeds have the potential to be used as natural coagulants.

Keywords: *Carica papaya*; kaolin; *Leucaena leucocephala*; *Moringa oleifera*; Turbidity

1. INTRODUCTION

Coagulation and flocculation are widely used as primary methods of water treatment. In the coagulation process, various chemical coagulants such as alum, ferrum, polyaluminium chloride (PAC) and polyferrous sulphate (PFS) are usually used. Although these coagulants work effectively, there are some drawbacks, such as a significant change in pH, inefficiency at low temperatures, and potential associated health problems (for example, Alzheimer's and dementia) (Srinivasan et al., 1999). Furthermore, these coagulants are relatively high in cost, and produce a high volume of sludge (Yin, 2010). Due to these drawbacks, efforts have been made to find alternatives, one of these being the utilization of natural coagulants, which may come from animals or plants. Plant-based coagulants have been used for 4000 years (Sutherland et al., 1990), and have now become an alternative water treatment, especially in rural areas (Mbogo, 2008; Pengchai et al., 2012). It is known that plant-based coagulants are safe for human health, low in cost and biodegradable, and that a lower volume of sludge produced compared to chemical coagulants (Yin, 2010).

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In this study, we utilize *M. oleifera*, leucaena and papaya seeds as plant-based natural coagulants to treat synthethic turbid water. They are native plants that can be easily found in Indonesia. *M. oleifera* has been studied intensively by various researchers (Ndabigengesere et al., 1995; Ghebremichael et al., 2006; Amagloh & Benang, 2009; Beltrán-Heredia & Sánchez-Martín, 2009), however leucaena and papaya seeds are relatively unexplored. Yongabi et al. (2011) studied the utilization of whole papaya seeds as a natural coagulant to remove turbidity and fecal bacteria, while Al-Mamun and Basir (2016) used leucaena seed extract as a natural coagulant. However, the effect of various parameters in the utilization of papaya and leucaena seeds as natural coagulants to the coagulation process has not been studied to date. In this study, we explore the effect of seed size, dosage and pH on the removal of turbidity in synthetic turbid water. The qualitative characterization of the active coagulant agent was also studied using FTIR analysis.

2. EXPERIMENTAL METHOD

2.1. Preparation of Natural Coagulant

Dried *M. oleifera* seeds were obtained from a local market in Bandung, West Java, Indonesia, and the de-shelled seeds were used as a natural coagulant. Papaya fruits were also obtained from a local market in Bandung, while the leucaena pods were collected from trees nearby. The flesh of the papaya fruit and the outer part of the leucaena pods were removed to obtain the seeds. These were repeatedly washed to remove impurities, then dried using an oven at 110˚C until the water content was below 10%w. The obtained seeds were ground to obtain a powder with a mesh size of –0+40, –40+50, and –50+60. The powder was stored in a dessicator and used as a natural coagulant without any further treatment. The seed powder was also analysed using FTIR (Shimadzu FTIR 8400, KBr pellet method) to qualitatively observe the possible active coagulant component in the seeds.

2.2. Coagulation Tests

Synthetic turbid water was prepared by adding 1 gram of kaolin powder (technical grade) into 1 L of tap water, followed by mixing at 60 rpm for 20 min to ensure hydration of the kaolin and subsequent formation of colloids, resulting in a solution with a turbidity of 862.6±46.6 NTU. The pH of the solution was adjusted using 1 M HCl or NaOH solution, prior to the jar test experiments. The coagulant seed powder was added at various dosages, and rapidly mixed at 200 rpm for 1 minute, followed by slow mixing at 60 rpm for 30 minutes. These mixtures were then left undisturbed for 1 hour to settle. The turbidity (NTU) of the solution before and after coagulation was measured using a turbiditimeter (Eutech Instruments Turbiditimeter TN-100), and the removal was calculated using Equation 1.

$$\%removal = \frac{initial\ turbidity - final\ turbidity}{initial\ turbidity} \times 100\% \quad (1)$$

The dosage and pH were varied in the experiment, following the design matrix presented in table 1.

Table 1 Variation of dosage and pH

Variable	-α	-1	0	+1	+α
Dosage (g/L)	0.29	0.5	1.0	1.5	1.71
pH	8.58	9.0	10.0	11.0	11.4

3. RESULTS AND DISCUSSION

3.1. FTIR Analysis

The FTIR analysis was made to obtain the active functional groups at the surface of the natural coagulants. The spectra presented in Figure 1 show strong and sharp peaks at 3600–3200cm^{-1}, indicating –OH and N-H overlapping stretching vibrations (Betatache et al., 2014), symmetrical, asymmetrical stretches, and bends of C-H around 2950, 2850, and 1460cm^{-1} respectively (Fatombi et al., 2013; Shak & Wu, 2014; Janakiraman & Johnson, 2015). The C=O functional group of carbocylic acids and esters was observed around 1780–1710cm^{-1} (Fedala et al., 2015), and peaks at 1650, 1540, and 1240 cm^{-1} belong to the primary, secondary and tertiary amides, respectively (Wang et al., 2009; Fatombi et al., 2013). The –OH bond in the phenolic structure was observed at a peak of 1270 cm^{-1} (Bodirlau & Teaca, 2009), with peaks between 1440–1398 cm^{-1} indicating –OH in the plane bends of the carboxylic acid structure (Pearson & Slifkin, 1972). C-O-C glycosidic bonds of polysaccharides were also observed around 1050-1170 cm^{-1} (Lammers et al., 2009). These FTIR results show that all the seeds contain active agents that can be used as natural coagulants, especially proteins. It is known that lectin in *M. oleifera* is the active agent in its coagulation (Okuda et al., 2001; Madrona et al., 2010), while the active agents of papaya and leucaena are still unknown. However, it is known that papaya seeds contain 27.8%w of protein (Marfo et al., 1986a), of which a 23.7%w fraction is soluble in 5% NaCl (globulin) and only 10.9% is water soluble (albumin) (Marfo et al., 1986b), while leucaena contains 57.8%w (dry basis) of protein (Sethi & Kulkarni, 1994), with a 44.9% fraction being globulin, and 29.3% albumin (Sethi & Kulkarni, 1993).

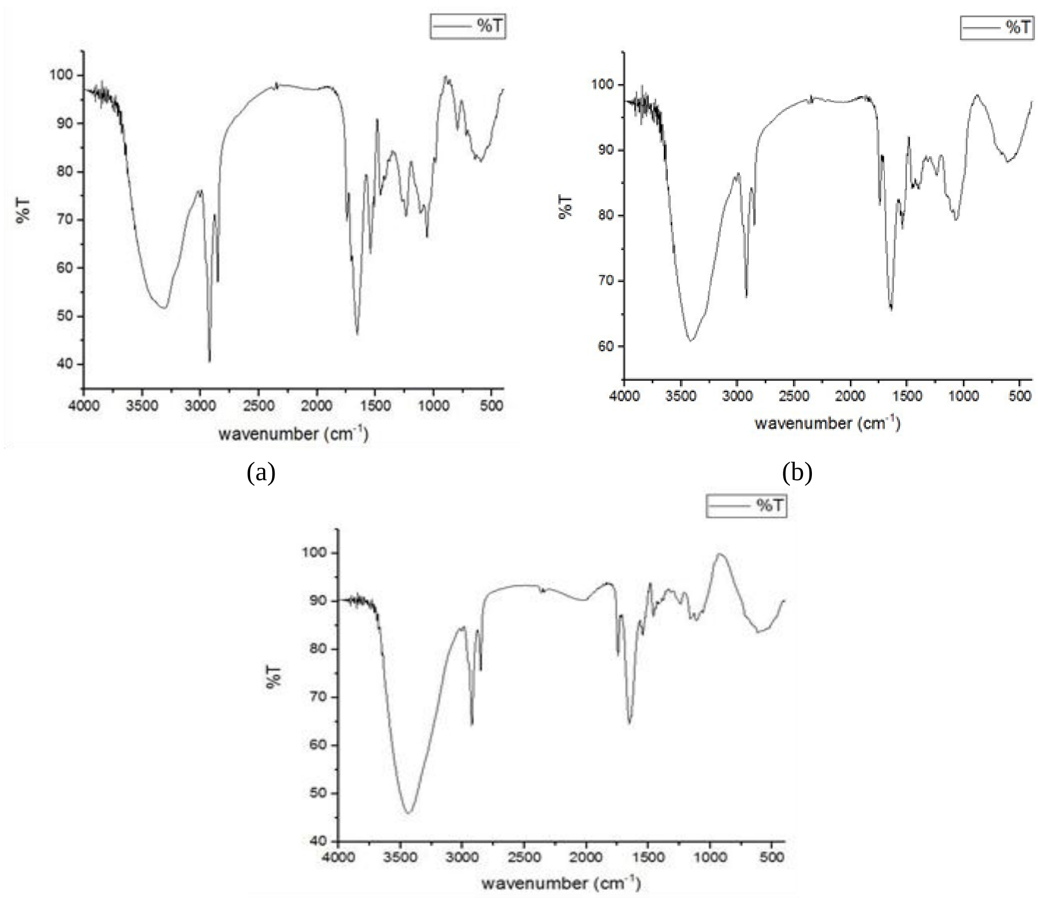

Figure 1 FTIR spectra of: (a) *M. Oleifera*; (b) Leucaena; and (c) C. Papaya seeds

3.2. The Effect of Seed Size on Turbidity Removal

The effect of seed size on its performance as a natural coagulant is presented in Figure 2. It can be observed that in the case of *M. oleifera*, its unground (whole) seeds gave the highest turbidity removal, while for the papaya seeds, the highest removal was observed at –40+50 mesh, and leucaena seed size had no effect on turbidity removal. It is known that particle size plays an important role in extraction; the smaller the particle size, the greater the number of compounds that can be extracted (Asep et al., 2008; Gião et al., 2009; Sari & Velioglu, 2011). However, with smaller seed sizes, compounds other than the active coagulant agent can also be extracted to water, thus contributing to the turbidity measured. We speculate that some compounds, such as carbohydrate and fibre, were dispersed to the water with the small seed size, which led to lower turbidity removal. For further dosage and pH study, seed size was used at its highest level of turbidity removal, which is whole seeds for *M oleifera*, and –40+50 mesh for leucaena and papaya.

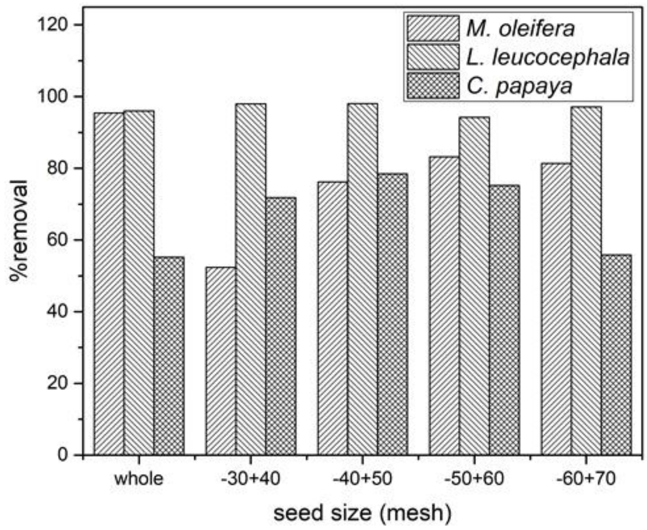

Figure 2 % removal of turbidity with various seed sizes

3.3. Effect of Dosage and pH on Turbidity Removal

The effect of the dosage (g/L) of various seeds and pH on turbidity removal is presented in Figure 3. It can be observed that the dosage and pH did not have any effect on the turbidity removal of *M. oleifera* (Figure 3a.), although there were some changes in turbidity removal when leucaena and papaya were used as the natural coagulants (Figures 3b and 3c). About 98% turbidity removal was obtained by *M. oleifera*, regardless of the dosage and pH of the solution, while leucaena gave 87.6% removal (1g/L, pH 10) and papaya 93.6% (1g/L, pH 10).

The results obtained for the dosage and pH variation in turbidity removal in this study are in accordance with those obtained by various other researchers. The results obtained by Okuda et al. (2001) showed that *M. oleifera* had high coagulant activity at pHs above 8, with constant coagulant activity up to pH 12 (Okuda et al., 2001). Similar results were also reported by Šćiban et al. (2005), showing that at pHs higher than 10, various legumes displayed high coagulant activity (Šćiban et al., 2005).

With regard to the dosage effect, it can be seen that the *M. oleifera* dosage had no effect on turbidity removal, whereas there was an optimum dosage level for leucaena and papaya. The effect of *M. oleifera* dosage was reported by Pritchard et al. (2010); beyond the optimum coagulant dosage, the coagulation effect became less effective, thus constant % removal was obtained (Pritchard et al., 2010). Overdosage of coagulant tends to lead to a colloid restabilization effect (Choy et al., 2015); however, this effect is not observed in *M. oleifera* (Ndabigengesere et al., 1995). Different trends were observed with both leucaena and papaya (Figures 3b and 3c). At low coagulant dosage, turbidity removal was low due to insufficient coagulating agent (Choy et al., 2015); however, overdosage could lead into colloid restabilization. This effect is shown in Figures 3b and 3c; with 1.5g/L coagulant dosage turbidity removal decreases.

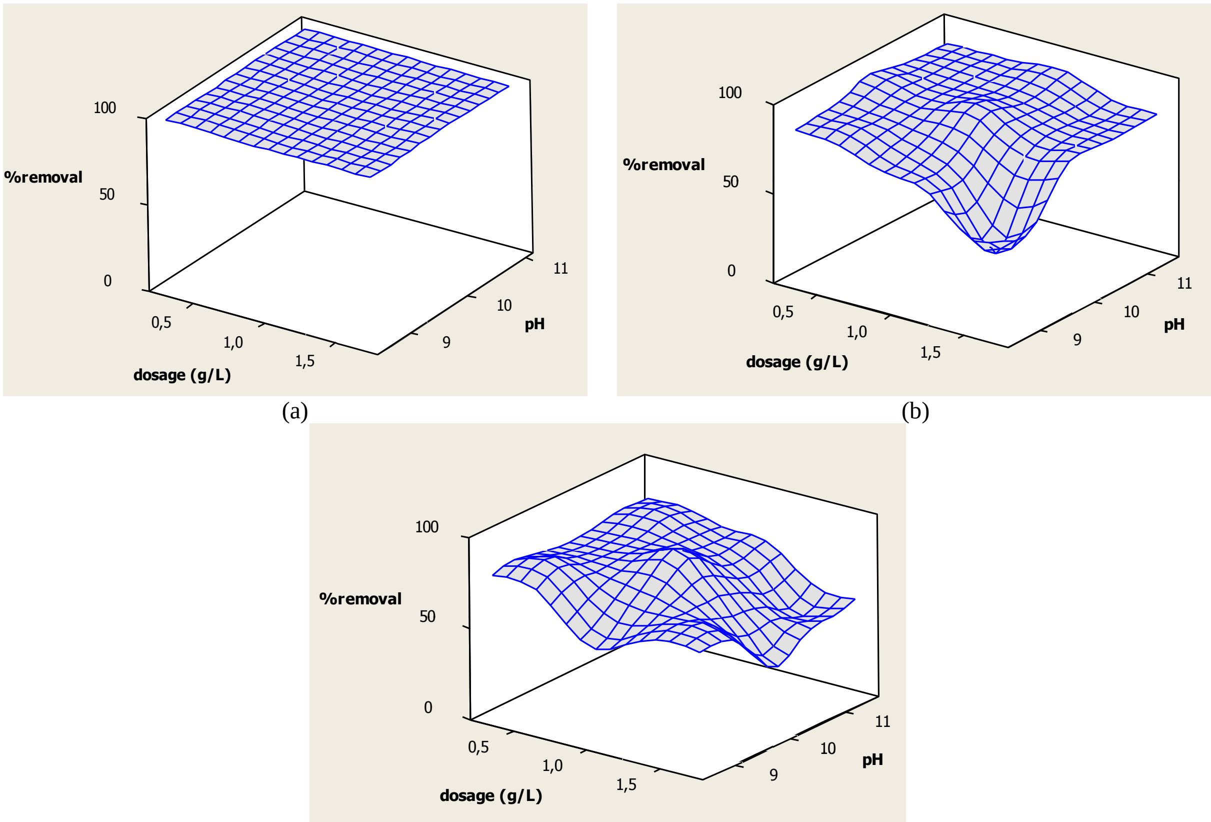

Figure 3 Effect of dosage (g/L) and pH on turbidity removal: (a) *M. Oleifera*; (b) leucaena; and (c) papaya

The protein in the seeds was the active coagulant agent in this study. It is known that protein is an amphoteric molecule, which is highly influenced by pH; at a pH above its isoelectric point, the protein molecule will have negative surface charge. The lectins in *M.oleifera* have an isoelectric point of 10 (Ndabigengesere et al., 1995). Leucaena protein is high in glutamic acid, leucine and lysine amino acids (Ekpenyong, 1986; Ahmed & Abdelati, 2009), with isoelectric points of 3.22, 5.98, and 9.74 respectively (Lundbland & Macdonald, 2010). Papaya seed protein is high in glutamic acid, threonine, proline and leucine (Marfo et al., 1986b), with isoelectric points of 3.22, 5.60, 6.30, and 5.98, respectively (Lundbland & Macdonald, 2010). The isoelectric point of kaolin is known to be at pH 2.8 (Ndabigengesere et al., 1995), therefore in the experimental pH range, both colloid and protein had negative surface charges. Consequently, we speculate that adsorption followed by interparticle bridging was the possible mechanism in this study.

A comparison between this work with other studies is shown in Table 2. It can be seen that our results are comparable with others, although the seeds were used directly, without extraction. Even *L. leucocephala* displayed higher turbidity removal, compared with results obtained by Al-Mamun and Basir (2016). Further study of the effect of various turbidities, sludge volume and the effect of salt extract for leucaena and papaya seeds is needed.

Table 2 Turbidity removal using various natural coagulants

Initial turbidity (NTU)	Coagulant	% removal	Reference
450	*M. oleifera* seed extract (NaCl 1M)	99.8	Nkurunziza et al. (2009)
296	*C. papaya* seeds	96.8	Yongabi et al. (2011)
250	*P. ovata* purified seed extract	95.6	Ramavandi (2014)
500	*M. oleifera* seed extract (NaCl 0.5M)	99	Muthuraman and Sasikala (2014)
	P. vulgaris seed extract (NaCl 0.5M)	95	
	S. potatorum seed extract (NaCl 0.5M)	90	
319	*L. leucocephala* seed extract (NaCl 1%)	76	Al-Mamun and Basir (2016)
862.6±46.6	*M. oleifera* seed powder	98.8	This study
	L. leucocephala seed powder	87.6	
	C. papaya seed powder	93.6	

4. CONCLUSION

The utilization of *M. oleifera*, leucaena and papaya seeds, which are indigenous plants in Indonesia, as natural coagulants has been studied in this research. From the FTIR spectra, it can be observed that all the seeds contained active coagulant agents, and could be used to treat turbid water. The coagulant dosage (g/L) and pH had no effect on the turbidity removal of*M. oleifera*, but some effect was observed when leucaena and papaya seed powder was used as the natural coagulant. The turbidity removal in this study was comparable with the results obtained by other researchers. Further study of the initial turbidiy effect, sludge volume after coagulation, and effect of salt extraction is needed.

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