

Communications:

Improving the Quality of Waste Water Containing Phosphate using *Moringa oleifera* Lam. Seeds

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Abstract: *Moringa oleifera* Lam. seed extract has been tested in removing phosphate in waste water. Coagulation has been carried out in order to study the mechanism this natural coagulant agent inside waste water. The decrease in total phosphate levels with the addition of Moringa 200 ppm and settling time of 90 minutes can reduce the concentration of total phosphate 23.119 ppm to 15.051 ppm, reduce about 27.82% and 13.047 ppm orthophosphate to be 9.852 ppm or reduce about 29.87%. Affectiveness of *M. oleifera* Lam. on pH 2 can decrease phosphate's concentrate as 52.15 % ppm and orthophosphates as 56.70 % ppm. Decreases of phosphate's concentrate in wastewater is caused by electrostatic force between groups of NH_3^+ in *M. oleifera* Lam. with H_2PO_4^- in wastewater. In this case, it is caused by protein content of *M. oleifera* Lam. that is supported by FTIR. The benefit of *M. oleifera* Lam. as phosphate coagulant is the witness of Allah's greatness. Allah's creation would not be wasted even if it is small like *M. oleifera* Lam. seeds.

Keywords: Coagulation-flocculation, *Moringa oleifera* Lam., Natural flocculants, Phosphate

1. Introduction

M. oleifera have several names according to the several States such as Arabic: rawag, Assamese: saijna, sohjna, Bengali: saijna, Burmese: daintha, dandalonbin, Chinese: la ken, English: drumstick tree, horseradish tree, ben tree French: moringe à graine ailée, morungue, Gujarati: midhosaragavo, saragavo, Hindi: mungna, saijna, shajna, Kannada: nugge, Konkani: maissang, moring, moxing, Malayalam: murinna, sigru, Marathi: achajhada, shevgi, Nepali: shobhanjan, sohijan, Oriya: saijina, Portuguese: moringa, moringueiro, Punjabi: sainjna, soanjna, Sanskrit: shobhanjana, sigru, Sinhalese: murunga, Spanish: ángela, ben, moringa, Swahili: mrongo, mzunze, Tamil: moringa, murungai, Telegu: mulaga, munaga, tellamunaga, Urdu: sahajna (Roloff *et al.* 2009)

M. oleifera Lam. was known in Indonesia as kelor (java), *Marongghi* (Madura) a vegetable crop that has been cultivated for a long time. The leaves are compound, pinnate double and small rounded. The flowers are yellowish-white. The fruit is long and angled-corner on its side. *M. oleifera* Lam. tree is often used as a hedge or a supporter for betel pepper plant. Today *M. oleifera* Lam. was acknowledge around the world as nutritious crop, medical plant and also absorbent and coagulation water treatment. Based on the test results of phytochemical that the chemical content of *M. oleifera* Lam. leaves showed alkaloid compounds and steroid / triterpenoid (Rizka and Nawawi 2005). The seeds contain 19 to 47 percent. It was known commercially as 'ben oil', it is similar to olive oil rich in palmetic, stearic, behmic, oleic acids, and is used for human consumption, cosmetics and soaps (Roloff *et al.* 2009).



Fig. 1. *M. oleifera* Lam. Plants

As well as medicine plant, *M. oleifera* Lam. also can be used as an absorbent and coagulation. The seed extract contains very interesting behavior in removing anionic surfactants from surface water. A very high efficiency is observed in all of the studied cases, so it presents a promising future as water treating agent. Regarding the influence of the coagulant dose, it is observed that a maximum surfactant removal is achieved with relatively low coagulant amounts (ca. 100 mg L⁻¹), (Martin and Heredia 2009).

In large scale treatment plants aluminum sulfate is used as conventional chemical coagulant. As an alternative to conventional coagulants, *M. oleifera* Lam. seeds can be used as a natural coagulant (primary coagulant) in household water treatment as well as in the community water treatment systems. When the crushed seeds are added to raw water, the proteins produce positive charges acting like magnets and attracting towards the predominantly negatively charged particles such as clay, silk, bacteria and other toxic particles in water. The flocculation process occurs when the proteins bind the negatives charges forming flocs through the

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aggregation of particles which are present in water (Schwarz 2000).

Hidayat (2003) shows the effectiveness of *M. oleifera* Lam. seeds biofloculant wastewater treatment process for pulp and paper industry. The measured parameters were the time of deposition, the value of color, turbidity, total solids, Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD). Hidayat (2006) again explained that *M. oleifera* Lam. seeds can be used as water purifiers because it contains positively charged protein as cationic polyelectrolyte better than alum (it is aluminium sulfate or tawas) as not to affect the pH and conductivity. Sludge produced by coagulation with *Moringa* is less in volume than the chemical sludge produced by alum coagulation.

This research aims to study the fundamental aspects of the main characteristics of functional groups of *M. oleifera* Lam. or *Moringa* seed powder, and their roles in the coagulation process. This study is also expected to reveal the ability of *Moringa* seeds that contain positively charged protein with an opposite charge compounds (with the anionic phosphate samples in the hospital sewage samples, which is expected to increase the utilization of *Moringa's* seeds which is part of the potential biodiversity of Indonesia.

Phosphate is a micronutrient compound of phosphorus. Phosphate in concentrations that exceed the quality standards will disrupt the existing balance of nitrogen compounds in the water, toxic to microorganisms and corrosive (Fachrul *et al.*, 2006). The concentration of phosphate in the water is usually determined by the number of water hyacinth plants growing on the surface of the water, but the water hyacinth on the surface of the water can block the sunlight needed microorganisms or green plants for photosynthesis, thus damaging the balance of water and water bodies of oxygen deprivation needed by water biota.

2. Materials and Methods

2.1. Preparation of *Moringa* Seed

The matured *moringa* fruits was prepared and peeled to obtain the white *moringa* seed. The seed was planted in porcelain pot and then stored in jars and sealed.

2.2. Sampling

The sample used in this study is a public hospital wastewater from Dr. Saiful Anwar Malang's hospital. Samples were taken using a glass rinsed with a solution of 0.01 N HCl (Clesceri *et al.* 1989). Preservation of samples were done by placing the samples at $4\text{ }^{\circ}\text{C} \pm 2\text{ }^{\circ}\text{C}$ (Hadi, 2005) or can be placed in the freezer (Clesceri *et al.* 1989).

2.3. Dose Determination of Optimum Coagulant Sedimentation and Coagulation Time of Phosphate Using *Moringa*

Moringa seed powder is made with various concentration of 200 ppm, 250 ppm, 300 ppm, 350 ppm, and 400 ppm respectively, coagulant was further diluted with 100 ml of sample. Samples interaction with coagulant *Moringa* seeds were carried out by the following procedure: *Moringa* seed powder was placed on a glass watch and added with a little sample that need to be analyzed, then stirred until a solution is

become white. Thus, inserted samples containing *moringa* seeds into the vial will contain samples that need to be analyzed. Samples containing *moringa* powder was stirred rapidly for 0.5 minutes and then stirred slowly for 5 minutes. The solution was allowed to settle with a variety of time *i.e* 15, 30, 60, 90, and 120 minutes. After that, 12 mL total phosphate and orthophosphate was pipet and were analyzed using a spectrophotometer HACH 4000. Samples were taken again at 25 ml and are used to measure the conductivity and pH of the sample. This treatment was also done on the control solution.

2.4. Dose Determination of Optimum Coagulant Sedimentation and Coagulation Time of Phosphate Using *Moringa* Seed

The pH Sample was measured by variations of 2, 3, 4, 5, and 6 with the addition of 0.1 N H₂SO₄ or 0.1 N NaOH, and then added with powdered of *Moringa* seeds with optimum dose. Interaction with *moringa* seeds were carried out by the following procedure: *Moringa* seed powder with optimum dose was placed on a glass watch and added a little sample of the conditions of pH 2, and stirred until the solution is become white. Samples containing *Moringa* seeds were put into a glass beaker containing the sample to pH 2 conditions to be analyzed. Samples containing *Moringa* were rapidly stirred for 0.5 minutes, then slowly stirred for 0.5 minutes. The solution was allowed to settle with optimum settling time. Each solution of total phosphate and orthophosphate was pipet at 12 ml and then analyzed using a spectrophotometer HACH and 4000 with Stano Chloride method respectively. This treatment was repeated with the same procedure with sample variation at 3, 4, 5, and 6 pH conditions.

2.5. Data Analysis

The results will be presented in graphic form and statistical test to determine the optimum dose and timing of deposition of *Moringa* seeds. Statistical tests were performed at two stages which are preliminary test and advanced test. Preliminary test was conducted by analyzing the F-test, RAK, RAL, and ANOVA, but this study employed two-way ANOVA test. If the results of the preliminary test found significant difference, then the test continued by BNT test to determine the slightly significant difference the influence of variations in dose and time deposition to decrease the phosphate concentration. The statistical result of deposition dosing and timing used as the optimum dose and optimum time deposition in determining optimum pH. *Moringa* seed powder characteristics determination was done by using the FTIR.

3. Results and Discussion

3.1. Samples Made Using Coagulation Coagulant of *Moringa* Seeds

The aim of coagulation is to compare the results between the coagulation and artificial samples of wastewater of Dr. Saiful Anwar Malang's hospital, because the composition of the wastewater is very complex. Table 1. shows the impact of *Moringa* seeds on phosphate concentration decreased with time variation of the deposition of 0, 15, 30, 60, 90, and 120

Table 1. Observations Samples Made Coagulation (16 ppm phosphate solution with a concentration of 200 ppm *Moringa*).

Time	Fosfat Average	Fosfat Coagulation	% decrease	conductivity average	pH average
0	15.935	0	0	0.98	7.01
15	15.484	0.45	2.83	0.98	6.93
30	14.942	0.92	5.8	0.94	6.87
60	14.31	1.55	9.79	0.93	6.82
90	14.04	1.86	11.69	0.91	6.79
120	13.282	2.54	16.08	0.85	6.71

minutes. Based on Table 1, it can be stated that the dose 200 ppm *Moringa* seeds can reduce phosphate concentrations of 2.54 ppm or 16.08%. The decreasing is relatively small because the concentration of *Moringa* seed as coagulant is less effective if the sample has a low value of turbidity (cloudiness) (Katayon, *et al.*, 2004: 150). Meanwhile decreasing in concentration due to the reduction of destabilizing colloidal repulsion force between phosphate ions and the process of inter-particle bridges. Ndabingengesere (1995: 708) and Katayon (2004: 151) states that *moringa* seed coagulation mechanism is dominated by the absorption and charge neutralization. The greater the energy of attraction, then the distance between the particles decreases the charge resulting in double diffusion layer depletion. The more neutral colloidal charge, it will form a flocs and finally settles.

Decreasing in phosphate concentration is supported by the decline in the value of the conductivity of the sample to 0.85 mS / cm indicating a reduction of phosphate ion mobility. *Moringa* seed granting phosphate ions causes charge each other to move in the opposite direction and then bind, so the mobility of ions is moving slowly. If the ion mobility decreases, the conductivity is also reduced (Atkins 1990; 313).

Solution conductivity is strongly influenced by the presence of large concentrations, ion movements generated and the temperature at the time of measurement (Hidayat 2006; 143). Atkins (1990) also states that the conductivity depends on the mobility of the ions, the greater the mobility of ion conductivity.

Radojevic (1999) states that H^+ , Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} , and HCO_3^- has a great influence on the value of conductivity, whereas ions such as Fe^{2+} , Fe^{3+} , Mn^{2+} , Al^{3+} , NO_3^- , HPO_4^{2-} , and $H_2PO_4^-$, and $H_2PO_4^-$ have little effect on the value of the conductivity of the water solvent.

Moringa seed causes a decrease in pH and becomes more acidic around 7.01 turns into 6.71. It is strengthened by Hidayat (2006) who states that the pH of the Lematang river water samples changed from 7.03 to 6.84 after adding 30 ppm coagulant of *Moringa* seed. The pH changes due to the carboxyl group of amino acids in *Moringa* seeds that release H^+ ions in acidic conditions. Katayon (2004) states that a relatively small decrease in pH occurred after coagulation of *Moringa* seeds between pH of 6.5 to 7.0. This is due to the

function of hydrogen ions (H^+) of a weak acid in balancing *Moringa* seeds samples with hydroxide ions.

3.2. Coagulating Using *Moringa* seeds

3.2.1. Coagulating activity of shelled *Moringa* seeds at different concentration

The results for determination of the optimum dose and timing of *Moringa* seeds deposition in lowering levels of total phosphate is shown in Fig. 2.

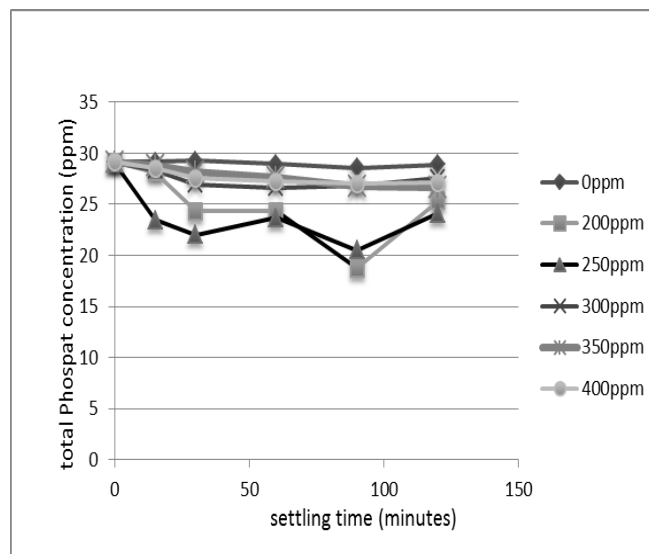


Fig. 2. Coagulating activity of shelled *Moringa* seeds at different concentration activity to decrease Total Phosphate in hospital wastewater.

Data were obtained at a dose of 200 ppm have optimum settling time at a minute of 90 with a decrease of 8.068 ppm or 27.04%. At a dose of 250 ppm, optimum settling time occurred in the 90th minute with a decrease of 8.754 or 30.15%, whereas a dose of 300 ppm optimum deposition occurred in the 90th minute with a decrease of 3.862 ppm or 13.31%. At a dose of 350 ppm, optimum settling time occurs at a minute of 120 with a decrease of 4.296 ppm or 14.75%, while the dose of 400 ppm when precipitation occurs at a minute of 120 with a decrease of 3.7 ppm or 12.72%.

Fig. 2 and 3 shows a relatively small decrease in concentration due *Moringa oleivera*.Lam seeds as a coagulant is less effective if the sample has a value of turbidity is low, including the possible number of other compounds that participate sedimentation in hospital wastewater sample (Katayon *et al.* 2004).

Decrease in concentration due to the reduction of destabilizing colloidal repulsion force between phosphate ions and the process of inter-particle bridges. Ndabingengesere (1995) and Katayon (2004) states that *Moringa* seeds coagulation mechanism is dominated by the process of adsorption and charge neutralization. The greater the energy of attraction, then the distance between the particles decreases the charge resulting in double diffusion layer depletion. The more neutral colloidal charge, it will form a floc and finally settles.

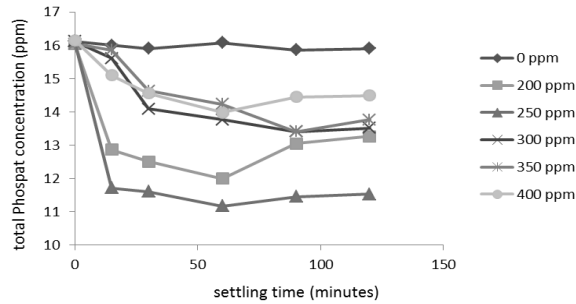


Fig. 3. Coagulating activity of shelled *Moringa* seeds at different concentration to decrease Total orthophosphate in hospital wastewater.

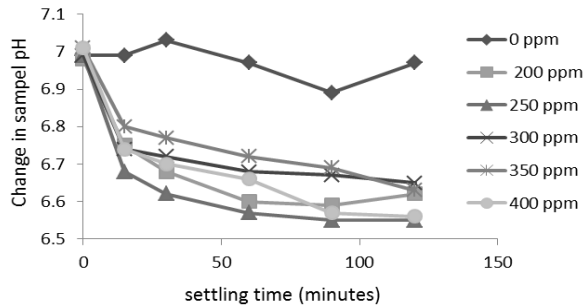


Fig. 4. Changes in effluent pH curves of hospital wastewater after interacted with *Moringa* seeds.

Fig. 2 and 3 shows that variations in dose and time *Moringa oleivera*. Lam settling influence on the concentration of orthophosphate and total phosphate but not significantly. Data analysis technique of determining the optimum dose and time of sedimentation conducted with a statistical test that includes two stages: a preliminary test phase and the test continued. Preliminary phase used is a two-way ANOVA test for the existence of two determining optimum conditions in the behavior of the phosphate concentration decreased. Data analysis followed by LSD test to determine the slightly significant difference influences from all over the dose variation (200 ppm, 250 ppm, 300 ppm, 350 ppm, and 400 ppm) *Moringa oleivera*. Lam seeds have a significant effect on phosphate concentration decreased. At doses of 200 and 250 ppm gives the greatest decrease in concentration, but the results of LSD test at a concentration of 200 ppm and 250 ppm did not give a real difference or that the effective dose is 200 ppm.

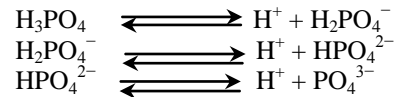
The results of two-way ANOVA showed that the variation of the deposition time the influence of the variation of the deposition time is done, while the BNT results show that in the 90th minute gave the real difference between the variation of the deposition time so time optimum deposition occurred in the 90th minute.

Increased phosphate levels again due because the downsizing boundary diffusion layer has reached the

maximum point, zeta potential of more than zero and van der Waals forces become weaker (Linggawati, 2002). Khalil and Aly (2001) in Linggawati (2002) states that the maximum flocculation occurs when the zeta potential price toward zero. Hidayat (1996) in Hidayat (2006) states that the addition of coagulant *Moringa* seeds excess can cause turbidity of raw water again clarified. The addition of excess *Moringa* seeds can cause increased distance between the charge and the repulsion force between like charges increase. This is also reinforced by Migo et al., (1993) in Novita (2001) which describes the adsorption of cation excess can cause deflokulasi or colloidal restabilization due to the repulsive force between the positively charged particles resist.

3.3. Effect of Dose Addition of *Moringa* seeds And Time Of The precipitation of pH changes

Fig. 4. show that pH changes after administration of *Moringa* seed for time variation but no significant precipitation. Changes in pH occur in about 6.6 to 6.9, while the initial pH around pH 7.0. Changes in pH is also influenced by *Moringa* seed for a protein that has a weak acid that can release H^+ ions in the medium of water, so many *Moringa* seeds will increase the pH of the sample (Katayon, et al. 2004). Ionization of phosphate in water medium at pH 7 with $H_2PO_4^-$, HPO_4^{2-} , and PO_4^{3-} forming compounds, which produces H^+ ions with a gradual, so that the sample pH acidic (Radojovic et al. 1999).



Changes in pH increased toward neutral pH, it is due to phenol and ammonia contained in the wastewater is more hospitals in the medium is alkaline water.

3.4. Effect of Dose Addition of *Moringa* Seed Deposition and Time of Changing Conductivity

Determination of conductivity performed to determine the effect of the addition of *Moringa* seed dose and time of deposition. The results of measurements of conductivity after interaction with *Moringa* seed presented in Fig. 5.

The conductivity of the samples has decreased although the decrease was not significant. Initial sample conductivity was 1.35 mS / cm, after interaction with *Moringa* seed of 200 ppm and 250 ppm with a 30-minute deposition conductivity values decreased to 1.34 mS/cm. It suggests that the reducing mobility of phosphate was coagulated with *Moringa* seeds, and not the only phosphate ion, the conductivity of the sample also affects very small decrease. Phosphate is a micro ion that can affect the value of conductivity, so the addition or reduction of phosphate levels little influence on the value of the conductivity of the sample.

Increasing the value conductivity exceeds initial conductivity is possible because the re-release coagulate electrolytes phosphate along with the addition of *Moringa* seed dose, but it is a polyelectrolyte *Moringa* seeds so that it will increase the number of ions in the sample and the conductivity value will increase.

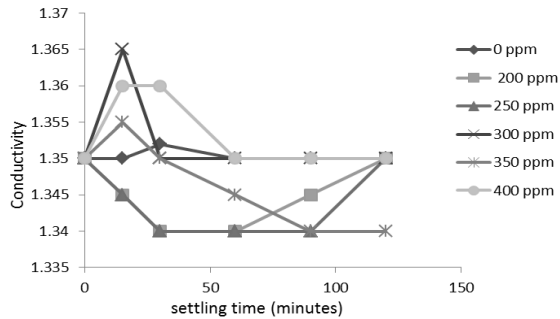


Fig. 5. Conductivity Change Curve Once interacted with Moringa in hospital wastewater.

3.5. Determination of Optimum pH Coagulation Phosphate Using *Moringa* seeds

Fig. 6 shows that at pH 6 phosphate concentrations decreased total 7.46 ppm or 25.62%. At pH 5 there is a decrease in total phosphate concentration of 11.39 ppm or equal to 40.53%, whereas at pH 4 a decrease of 11.89 ppm or equal to 42.98%. At pH 3 a decrease of 14.09 ppm or 48.47%, whereas at pH 2 a decrease of 14.92 ppm or 52.31%.

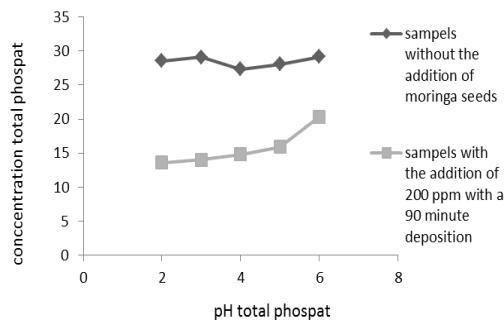


Fig. 6. Effect of pH Variation of Total Phosphate Concentration in Hospital Wastewater.

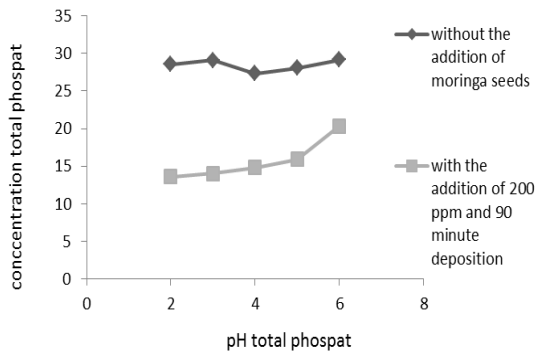


Fig. 7. Effect of pH Variation to orthophosphate concentrations in Hospital Wastewater.

The results of the influence of variations in pH to decrease in total phosphate concentrations are presented in Fig. 7. Fig. 7 shows that at pH 6 phosphate concentration decreased total 4.58 or 29.78%. At pH 5 phosphate concentration decreased by 5.36 or by 35.41%, whereas at pH 4 a decrease of 6.93 or by 46.22%. At pH 3 a decline of 8.27 or 56.07%, whereas at pH 2 a decline of 8.64 or 56.70%. The greatest decrease in phosphate concentration at pH 2, this is due to the formation of a cationic polyelectrolyte- NH_3^+ group at the highest pH 2 so as to precipitate phosphate greater, whereas in the range of pH 2-6 phosphate H_2PO_4^- shaped. Phosphate concentration at pH 3 to 6 a decline that is not very significant because of the pH-NH_3^+ group and declining possible- NH_2 group is increasing.

3.6. *Moringa* seed powder characteristics Using FTIR

Identification using infrared spectrophotometry (IR) aims to obtain information about the existence of a functional group of a molecule. It is because every functional group has a typical vibration region (Sastrohamidjojo 1992). *Moringa* seed powder spectra can be seen in Fig. 8 and 9, are as follows:

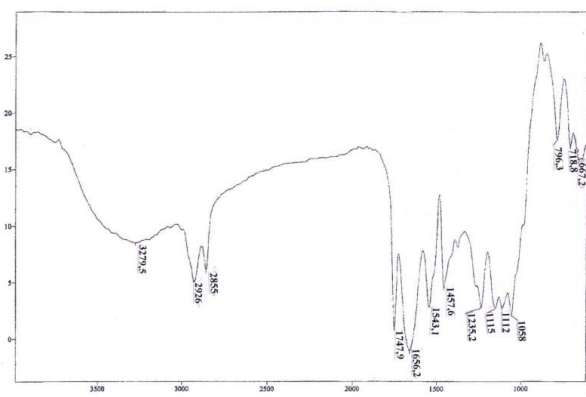


Fig. 8. *Moringa* seed powder spectra before interacted with Phosphate.

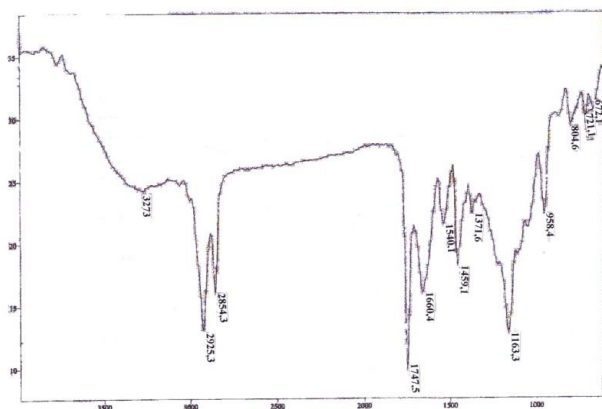


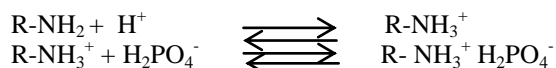
Fig. 9. *Moringa* seed powder spectra after interacted with Phosphate.

Based on the spectra at Fig. 9, there are differences in uptake between *Moringa* seed powder before and after coagulant used for phosphate. *Moringa* seed powder spectra before being used as a coagulant phosphate showed absorption at 3279.5 cm⁻¹ region which showed the presence of OH groups attached to the intermolecular hydrogen (single molecules or other molecules similar or different) with weak absorption intensity. Socrates (1994) vibrational states range 3550-3230 cm⁻¹, but little shift in the wave number becomes 3273 cm⁻¹. Carbonyl group C=O is shown by absorption at wavenumber 1747.9 cm⁻¹ ester group, the same is true of *Moringa* seed powder after interacted with phosphate, but a slight shift to 1747.5 cm⁻¹.

Moringa seed powder spectra before and after interaction with phosphate occurs much wavenumber shift include CH stretch of aromatic CH absorption wavenumber at 2855 cm⁻¹ shifted to 2854.3 cm⁻¹, C=O stretch of a secondary amide absorption wavenumber 1656.2 cm⁻¹ shifted to 1660.4 cm⁻¹, the deformation of the amide NH stretch, absorption wavenumber 1543.1 cm⁻¹ shifted to 1540.1 cm⁻¹, -CH₂-scissors vibration of alkanes on absorption wavenumber 1457.6 cm⁻¹ shifted to 1459.1 cm⁻¹, CH stretch out the field on the absorption wavenumber 796.3 cm⁻¹ shifted to 804.6 cm⁻¹, and the bending vibrations of the alkyl isothiocyanate (N=C=S) on the absorption wavenumber 667.2 cm⁻¹ shifted to 672.1 cm⁻¹. This shift is expected because these groups have interacted with the phosphate.

The parameter is to indicate which is the most interactions with phosphate *Moringa oleifera*. Lam seeds is physical absorption process. Absorption is reversible and fast, but it also indicated chemical absorption occurs with the formation of the new vibrations *Moringa* seed powder as-CH stretch vibration effect on the uptake of symmetry deformation wavenumber 1371.6 cm⁻¹, due to stretch vibrations P=O compounds suspected of phosphate on the sorption wave 1163.3 cm⁻¹, and the range of PN presumably from PNC vibration absorption wavenumber at 958.4 cm⁻¹. The chemical absorption occurs with the formation of chemical compounds that bond that occurs more powerful, it is also confirmed by Ndabingengesere (1994) who states that most of the seeds of *Moringa* coagulation process is are the process of adsorption and charge neutralization.

Moringa seed powder spectra after interaction with phosphate showed an increase in intensity as the vibration of -CH₂- asymmetry and aromatic CH stretch due to the phosphate attached to the seeds of *M. oleifera* Lam. Spanning P=O are also present in the spectra after interacted with *Moringa* seed suspected of phosphate compounds on the wave absorption and 1163.3 cm⁻¹ range PN suspected of vibration absorption wavenumber PNC at 958.4 cm⁻¹, data This suggests that phosphate binds to the N, nitrogen, presumably from NH₃⁺ from *Moringa* seed proteins. The reaction occurs as follows:



Description: A group of other *Moringa* seeds.

4. Conclusions

1. The optimum dose of phosphate wastewater coagulation general hospital Dr. Saiful Anwar Malang using *Moringa* seed that is 200 ppm greater dose of *Moringa* seeds were added did not provide better results, this is due to a weak bond between phosphate and *Moringa* seed that phosphate detaching.
2. The greatest decrease in phosphate concentrations obtained at the time of deposition of 90 minutes, the longer the settling time or contact time with phosphate, *moringa* seeds did not provide more phosphate concentration decreased, this is due to the weak bond between the phosphate and the amount of electrolyte *moringa* seeds besides anionic phosphate coagulation participate.
3. Decrease in phosphate concentration was greatest at pH 2, this is due to the NH₃⁺, which has been formed to or greater than the NH₂ group at acidic pH and at pH 2, phosphate H₂PO₄⁻ forming compounds.

5. Acknowledgments

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