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EFFECTIVENESS OF *MORINGA OLEIFERA* AS A NATURAL COAGULANT AND ADSORBENT IN LEACHATE TREATMENT

ERINA RAHMADYANTI[1]*, NINIK WAHJU HIDAJATI[1] AND MAS SURYANTO HADI SAPUTRO[1]

[1]*Departement of Civil Engineering, Faculty of Engineering, Universitas Negeri Surabaya, East Java, Indonesia*

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ABSTRACT

Leachate is wastewater from garbage heaps that has the potential to pollute the environment if it is not treated. The physical and chemical technologies that are often used are coagulation flocculation and adsorption. Both of these technologies use synthetic coagulants and adsorbents to reduce pollutants in leachate. The use of synthetic coagulants and adsorbents is harmful to the environment and human health. This study aims to determine the effectiveness of *Moringa oleifera* as a natural coagulant and adsorbent in the coagulation flocculation and adsorption in leachate treatment. The result shows that *Moringa oleifera* as a natural coagulant was able to remove COD, TSS, and heavy metal respectively by 62%, 94%, and 88% compared to alum, namely 64%, 88%, and 78%. The effectiveness increased with the combination of adsorption using fly ash which was able to remove COD, TSS, and heavy metal to 74.2%, 99.5%, and 99.6% respectively compared to 76.6%, 96.3%, and 97.4% alum. The addition of *Moringa oleifera* needs to be considered because it makes the environment tend to be alkaline even though it is still in the normal range of 6-8. Particularly for COD, although it has high efficiency but does not meet government regulatory standards, therefore it is recommended to combine it with biological treatment in future studies.

KEY WORDS : Leachate, Environment, Coagulant, Adsorbent, *Moringa oleifera*

INTRODUCTION

Sanitary landfills are the most common method used around the world to treat waste because of their low operating costs (Wang *et al.,* 2018). The by-product of sanitary landfill is leachate which contains a lot of organic and inorganic contaminants. Leachate is a liquid that is formed from a biological degradation process, black in color and very smelly. Leachate generally contains high levels of organic (hydrocarbons, humic acid, sulfuric, tannic and gallic) and inorganic compounds (sodium, potassium, calcium, magnesium, chlorine, sulfate, phosphate, phenol, nitrogen and heavy metal compounds) due to high levels of COD and ammonia in leachate (up to thousands of mg/l) (Kanmani and Gandhimathi, 2013; Sivakumar, 2013).

The characteristics of leachate vary depending on the composition of leachate, volume, and the content of biodegradable matter which depends on its age (Samadi *et al.*, 2010; Raghab *et al.,* 2013). The pollutant content in leachate is equivalent to 100 times the pollutant content in domestic waste (Chen *et al.,* 2018). Leachate which is discarded without being treated can seep through the soil and underground layers to become a potential source of surface water and groundwater contamination causing pollution in receiving water (Abdul Aziz *et al*., 2011) (Kamaruddin, 2015; Dia *et al*., 2018).

Leachate treatment is based on its composition and characteristics (Aziz *et al*., 2018). Biological treatments are generally environmentally friendly but only effective for young or newly formed leachates. Meanwhile, the physico-chemical method is more effective for treating older leachate which contains various types of metals and has high COD, ammonium and COD/ BOD ratios (Aziz *et al*., 2009). Leachate processing using coagulation flocculation technology using synthetic coagulants can reduce COD by 56% using Alum and 52% using ferric. Heavy metals can be reduced by 85% using

Alum coagulants and 68% using ferric coagulants (Malathi *et al*., 2016). Meanwhile, the use of activated carbon in adsorption and electrolysis technology has been proven effective in reducing suspended solids and heavy metal content in landfill leachates (Erabee *et al*., 2018). Leachate used in this study is leachate that has long been established (old leachate) where physical and chemical processing is expected to be able to reduce pollutants optimally.

The majority of physical and chemical methods have limitations on the expensive aspect of financing (Vieira *et al*., 2010). The physical-chemical processing methods that are often used are the coagulation and adsorption flocculation methods (Teh *et al.,* 2014). Adsorption is a method based on surface phenomena due to the accumulation of a species at the solid-liquid surface boundary (Daud *et al*., 2017). In adsorption activated carbon is used to adsorb certain gases or compounds or chemicals which have selective adsorption properties depending on the size or volume of pores and their surface area (Sumathi and Alagumuthu, 2014). Apart from adsorption, the coagulation flocculation method is widely used as a pretreatment because of its simplicity and convenience (Freitas *et al.,* 2015; Prasad and Rao, 2016). Flocculation is the process of bringing unstable particles together to form a larger mass so that they can be filtered or filtered at the next water treatment. Coagulation is the process of adding certain chemicals to bring together smaller particles that do not settle in the flocculation process (Gandhimathi *et al.,* 2013a).

In the coagulation flocculation process, inorganic coagulants and synthetic polymers are added, such as aluminum polychloride, aluminum sulfate, and ferric sulfate (Swelam *et al*., 2019). These various coagulants are able to reduce impurities, especially turbidity, toxic substances (organic and inorganic) and colloidal organic matter (Ying *et al*., 2011). However, it has several disadvantages such as the production of large amounts of non-biodegradable sludge and has a high level of toxicity (Dehghani and Alizadeh, 2016). The use of synthetic coagulants also increases the pH of the waters and is therefore considered a toxic environmental pollutant. In addition, synthetic coagulant residues in water can trigger Alzheimer's disease (Nouhi *et al*., 2019; Mataka *et al*., 2010). Likewise with the adsorption process, where activated carbon is widely used. Using activated carbon as an adsorbent is expensive (Mataka *et al*., 2010). Due to these reasons, currently there are many researches to study the use of natural coagulants and natural adsorbents, especially in developing countries.

Several studies have been conducted on the use of natural coagulants and adsorbents. Polysaccharide and protein content were identified as having potential as coagulants (Choi, 2015; Teixeira *et al.,* 2017). Other potentials possessed by natural coagulants and adsorbents are abundantly available, low cost, and non-toxicity (Tukki *et al*., 2016; Bongiovani *et al*., 2014). Several agricultural byproducts, such as coconut husks, corn husks and corn cobs, are reported to be heavy metal adsorbents (Mataka *et al*., 2010).

Among the natural coagulants and other adsorbents, *Moringa oleifera* is the most widely used. *Moringa oleifera* is native to India which consists of 14 species that grow well in hot climates and various types of soil. It is not surprising that *Moringa oleifera* is also widely available in Indonesia. *Moringa oleifera* has been widely used to treat wastewater (Vieira *et al.,* 2010) (Sivakumar, 2013). Previous research stated that *Moringa oleifera* as a coagulant/adsorbent was able to remove turbidity, COD, and dyes. *Moringa oleifera* was able to reduce turbidity to 98% and conductivity to 11%, whereas color removal was up to 75% with a combination of alum (Durairaj, 2017). *Moringa oleifera* seeds contain micro-mineral compounds, emulsions, glycerid acid, and polmiric acid, which function as chelates so they can attract metal ions and other particles (Vijayaraghavan and Sivakumar, 2011). The combination of coagulation flocculation and adsorption using *Moringa oleifera* can reduce COD and TDS optimally with COD removal reaching 84.5% and TDS up to 82.6% with a *Moringa oleifera* dose of 100 mg/l with a slow stirring time of 20 minutes and a fast stirring for 120 minutes (Ahmadi *et al.,* 2017).

The success of the method of coagulation flocculation and adsorption on leachate treatment and the potential use of natural coagulants and adsorbent to be the basis of this study. This study aims to determine the effectiveness of the use of *Moringa oleifera* as a natural adsorbent and coagulant on methods of adsorption and coagulation-flocculation compared with commercial adsorbent and coagulant on landfill leachate treatment.

MATERIALS AND METHODS

Material

Leachate landfill

The leachate used is formed from the process of decomposing waste from the Universitas Negeri Surabaya campus which is more than 5 years old.

Moringa oleifera seeds

This study used Moringa oleifera seeds obtained from Sumenep, Madura, East Java. *Moringa oleifera* seeds were crushed using a domestic blender and sieved using a 0.08 mm stainless steel sieve (Swelam *et al*., 2019). Then the *Moringa oleifera* seed powder was oven-dried at 60 °C for 36 hours. The oil content of *Moringa oleifera* seeds is extracted using the Soxhlet system. This process produces a by product which is then re-dried using an oven for 24 hours at 60 °C for evaporation of n-hexane used for oil extraction.

Coagulant preparation

In the first stage, the active ingredient extraction of *Moringa oleifera* coagulant is carried out by dissolving 5 g of dried *Moringa oleifera* powder in 500 ml of petroleum ether (1: 5 ratio) at a temperature of 40-60 °C using a magnetic stirrer for 120 minutes to dissolve the oil in the seeds. The solution was filtered using a 0.5 mm pore fiberglass membrane and the sediment is left in open air for overnight (Santos *et al*., 2012).

The protein was dissolved in deionized water (50 g each in about 200 mL water) using a magnetic stirrer for 60 minutes. The solution is then filtered to separate it from the solids until a clear solution is obtained (approximately 2-3 times). The protein is re-precipitated in the form of a sticky paste by adding ammonium sulfate to saturation. The paste was redissolved in deionized water and filtered again. The solution was dialyzed with high purity water using a cellulose membrane tube to remove ammonium sulfate. The protein purification process can be carried out further by adding the dialyzed solution to the packed carboxymethyl cellulose column. The bound protein is then released and eluted using 1 M NaCl solution to remove the remaining salt as well as obtain protein powder by freeze drying (Nouhi *et al.,* 2019).

Adsorbent preparation

The adsorbent used in this experiment is fly ash. Fly ash is coal burning waste that is stockpiled and has the potential to cause environmental problems (Syed Farman Ali *et al.,* 2015). The adsorbent preparation was carried out by washing fly ash using deionized water and then drying it for 24 hours. The dry fly ash is then ground and treated with 1 N nitric acid (1 N HNO_3), 1 N hydrochloric acid (HCl), and 30% w/v hydrogen peroxide for 24 hours to clean adhered organic impurities. Next, wash again with deionized water until a neutral pH is obtained and then oven for 2 hours at a temperature of 105 °C. Before use, fly ash must be activated in the furnace for 1 hour at a temperature of 500 °C. In the final step, sieving was carried out to divide the 3 size groups, namely less than 0.5 mm, 0.5-1.0 mm, and 1.0 - 2.0 mm. Adsorbent that is ready for use is stored in a vacuum desiccator until needed (Jarusiripot, 2014).

Experimental design

This study used two treatment groups as shown in Figure 1 namely 1) coagulation flocculation using alum and *Moringa oleifera*; and 2) a combination of coagulation flocculation and adsorption using alum and *Moringa oleifera*.

The coagulation flocculation process was carried out using a jar test at room temperature. In the initial stage, fast stirring was performed at 180 rpm for 1 minute followed by slow stirring at 50 rpm for 15 minutes. Furthermore, the deposition was carried out for 30 minutes. After the deposition process, the supernatant was withdrawn from the beakers and used for chemical analyzes and for adsorption study (Eri *et al*., 2018; Gaikwad and Munavalli, 2019). The same process is carried out using $Al_2(SO_4)_3$ or alum

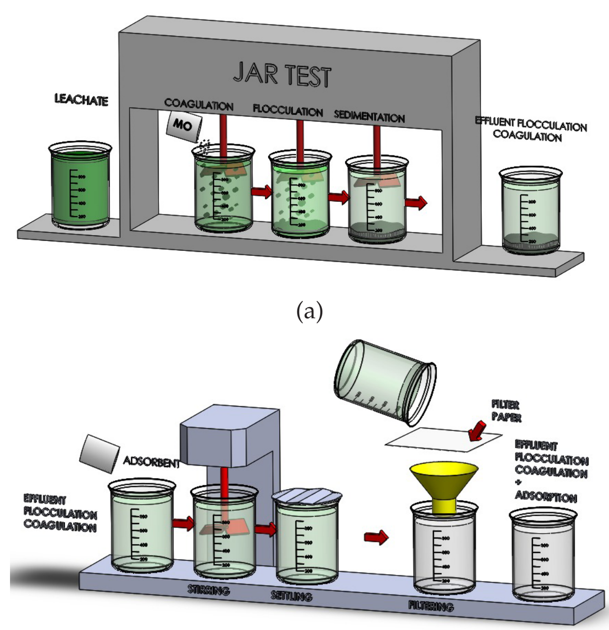

Fig. 1. (a) Flocculation coagulation proccess using coagulant *Moringa oleifera*; (b) Flocculation coagulation followed with Adsorption proccess using adsorbent *Moringa oleifera*.

as a comparison. The treated leachates were characterized by measuring the BOD, COD, TDS and heavy metal using the standard method (Güneº, 2014). The second treatment is the adsorption process carried out in batch conditions. The optimum conditions for the batch study were determined at room temperature, constant stirring rate for 105 minutes using 10 grams of media and 100 ml of leachate or 100 g/l of biosorbent concentration.

The adsorption process aims to remove every pollutant parameter contained in leachate. This test is performed using a 250 ml conical flask which has been filled with 100 ml of leachate. Furthermore, fly ashes were added as adsorbent treated with acid for activation with a contact time of 150 minutes. The agitation speed was changed from 50 to 200 rpm. All samples were assessed in triplicate under identical conditions, and the average was obtained. Then the samples were analyzed for the content of pH, COD, TSS and heavy metal using the standard method (Daud *et al*., 2017). The efficiency of each parameter was calculated using the equation: RE % = [(Co - Ce) / Co] × 100, where: Co and Ce are the initial and residual concentration of each parameter in water (mg/L), respectively.

Statistical analysis

Discriminant function analysis was performed to compare the effectiveness of coagulation flocculation treatment and the combination of coagulation and adsorption flocculation. Furthermore, to determine the removal efficiency of pH, COD, TSS, and heavy metals, a one-way ANOVA test was performed with a 95% confidence interval. SPSS 16 software with a significance level of $p < 0.05$ was used as a tool to perform this analysis.

RESULTS AND DISCUSSION

Characteristic of leachate landfill

The physical and chemical characteristics of leachate landfills before processing are shown in Table 1. The results show that the BOD_5/COD ratio <0.1, which means that the leachate is in the stabilized category. This value indicates the age of the landfill and the rate of biodegradation (Aziz and Ramli, 2018). Stabilized leachate has a low BOD_5 value and BOD_5/COD ratio (Al-Hamadani *et al*., 2011; Uygur and Kargi, 2004) which means that the level of biodegradability is low. A high TSS concentration (11,000 mg/l) indicates the presence of organic and inorganic solids (Al-Hamadani *et al*., 2011) (Azis *et al*., 2015) This high organic solid causes the color to become darker due to the presence of a substance similar to humic acid (Ishak *et al*., 2016) (Azis *et al*., 2015). However, leachate landfills have a pH of 7.68 or in the range 6-9, which means that biological life is possible (Noerfitriyani *et al*., 2018). The pH value of leachate is in line with previous research which states that stabilized leachate has a pH value that tends to be constant between 7.5 and 9 (Umar *et al*., 2010). The leachate conductivity value is not too high, namely 170 ì Mho/cm which indicates the amount of inorganic material is not too high (Gandhimathi *et al*., 2013). In leachate, heavy metal were detected even below the detectable limit. In stabilized leachate, where the pH tends to be higher than fresh leachate, it causes a decrease in the solubility of certain metals. Stabilized leachate samples have a lower concentration when compared to fresh leachate samples (Kanmani and Gandhimathi, 2013) (Sulaiman *et al.*, 2017).

Effect on pH

pH is the most important parameter in the coagulation flocculation treatment (Samadi *et al*., 2010). In the coagulation flocculation, alum is mostly used. The addition of alum as a coagulant in leachate treatment resulted in a decrease in pH from 7.68 initially to 7.52 as shown in Figure 2. This condition also occurred in the coagulation flocculation followed by the adsorption treatment, where the pH also decreased from 7.68 to 7.15. The addition of alum tends to lower the pH (Malecki-Brown *et al*., 2007). This results are in line with the previous study in which the addition of alum in muddy water with pH maintained at about 7 decreased pH near 6 after coagulation. Although there was a decrease in pH due to the addition of alum as a coagulant, it was still in a neutral condition (Malik, 2018).

The addition of *Moringa oleifera* seed as a natural coagulant, the pH increased from 7.68 to 8.02. The combination of adsorption treatment with the *Moringa oleifera* coagulant also gave results that were not much different, where the pH increased to 7.72. This increase in pH is caused by the receipt of protons from water by the alkaline amino acids contained in *Moringa oleifera* protein, resulting in the release of hydroxyl groups making the solution basic (Sapana *et al.*, 2012) (Dehghani and Alizadeh, 2016; Sapana *et al.*, 2012). In the adsorption process with fly ash, the pH tends to decrease due to the acidic nature of fly ash being neutralized by the dissolution of CaO and MgO where the reaction path is ultimately buffered by the absorption of carbon dioxide yielding a pH of 7 to 8 (Roy and Berger, 2011).

The addition of *Moringa oleifera* as a natural coagulant is able to maintain the pH in neutral conditions, allowing for further biological treatment. For the treatment of other pollutant materials that require certain conditions in an acidic or alkaline atmosphere in order to obtain a more optimum removal, other environmentally friendly materials can be added.

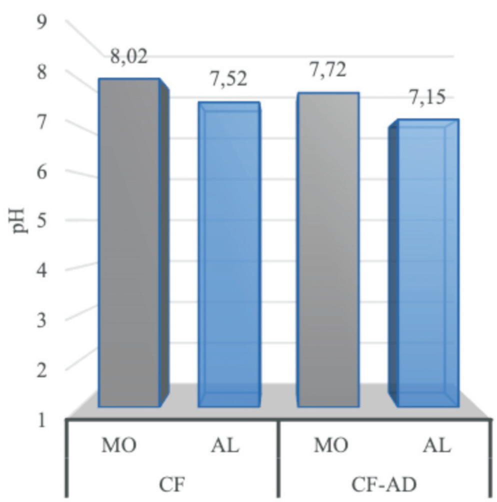

Fig. 2. Effect on pH

Effect on COD removal

COD describes the oxygen requirements needed to oxidize organic matter dissolved in water (Rahmadyanti and Febriyanti, 2020). The organic compounds contained in leachate are dominated by dissolved organic compounds that are difficult to biodegrade, characterized by a low BOD_5/COD ratio of only 0.11 and dark brown water, possibly including humic compounds. The COD value in this study indicates that leachate is classified in the methanogenic phase. The COD value of leachate is currently in the range for the methane fermentation phase (Ramprasad *et al*., 2019). The results showed that the use of *Moringa oleifera* as a coagulant was able to produce COD removal efficiency of 62% while alum reached an efficiency of 64% as shown in Figure 3. COD removal was due to load neutralization and pollutant melting on *Moringa oleifera* and alum. pH is maintained near neutral conditions (Gandhimathi *et al*., 2013b).

The addition of *Moringa oleifera* as a cationic polymer into the colloid particles with this negative charge will form a particle bridge between colloidal particles. The bridges are interconnected with each other so that enough mass is obtained to settle. The deposition of organic matter causes a decrease in the COD value. The decrease in COD is also caused by the antimicrobial properties of *Moringa oleifera*. Gram-negative and negative bacteria can be fluctuated by the protein present in *Moringa oleifera* seeds (Novita *et al.*, 2019). Another mechanism that may occur as a cause of the decrease in COD is the protonation of the *Moringa oleifera* amino group in solution making the positively charged *Moringa oleifera* act as a cationic polyelectrolyte. Because the particles in the leachate suspension are negatively charged, *Moringa oleifera* is very attractive as a coagulant by allowing the molecules to bind to negatively charged surfaces via ionic or hydrogen bonds. This in turn reduces or neutralizes the surface charge of the particles. Therefore, the destabilization of *Moringa oleifera* particles can be explained by a charge neutralization mechanism (Azis *et al*., 2015).

Alum has optimal efficiency in a pH range of 5–7. Even though the leachate used in this study, has a pH of 7.68 (tends to be alkaline), which causes alum not fully positive. Thus, they neutralize charge, absorb organic pollutants and solids, and consequently increase removal efficiency (Yang *et al*., 2010). Conditions that tend to be alkaline result in the removal process being not optimal due to the formation of neutralized Al hydroxide ($Al(OH)_3$) floc species, where charge neutralization and adsorption mechanisms do not occur to remove pollutants (Yang *et al*., 2010). The results showed that alum was able to achieve a removal efficiency of 64% which subsequently increased by 76.6% (Figure 3).

The addition of coagulants (alum and *Moringa oleifera*) that exceeds the dose. the optimum causes the decanter liquid to tend to rise. This indicates that the colloidal particles in leachate have undergone the same charge neutralization process or their iso electric point has been reached, so that the addition of more coagulant compounds causes some of the particles to experience restabilization because they receive a positive charge from the coagulant ions.

The adsorption combination caused the COD removal efficiency to be higher, namely to 74.2% with the addition of *Moringa oleifera* and 76.6% with alum (Figure 3). This illustrates that the material in

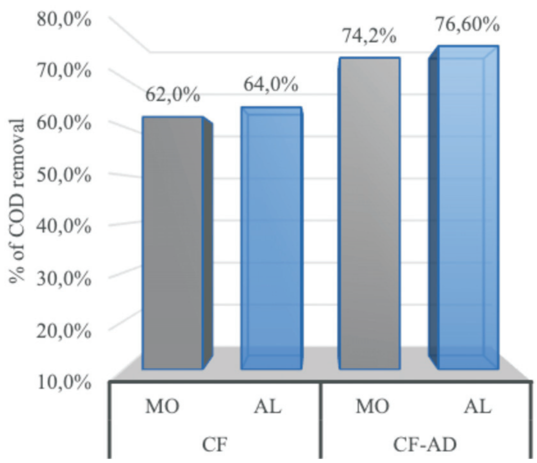

Fig. 3. Effect on COD removal

fly ash can capture organic compounds because it has high porosity. Its adsorption capacity depends on the interaction of leachate pollutants with fly ash adsorbents. In the adsorption process with fly ash, repulsion occurs between the colloidal organic matter and the negatively charged fly ash surface which causes an increase in COD removal efficiency (Gandhimathi *et al.*, 2013b).

The results of COD effluent obtained through the coagulation flocculation process and the combination of coagulation and adsorption flocculation still cannot meet the predetermined standards or quality standards. This is because there are too many organic and inorganic contents in leachate so that it requires further processing to remove larger COD levels. This may be due to several factors such as the relatively short deposition time (Novita *et al.*, 2019).

Effect on TSS removal

Total suspended solids are solid particles which include organic and inorganic materials that are soluble in water. High amounts of suspended solids in water bodies can cause sedimentation and disrupt aquatic life (Ali *et al.*, 2015). The results of this study as shown in Figure 4 shows that the addition of *Moringa oleifera* was able to remove TSS up to 94% with coagulation flocculation. TSS removal by *Moringa oleifera* was caused by destabilization of negatively charged colloids by cationic polyelectrolytes (Olanrewaju, 2018).

The mechanisms occurs is adsorption and charge neutralization, or adsorption and bridging unstable particles (Sotheeswaran *et al.*, 2011; Dehghani and Alizadeh, 2016). When the optimum dose of *Moringa oleifera* is exceeded, particle resuspension occurs. This is due to re-stabilization caused by reversal of the colloid charge due to adsorption or possible saturation of the polymer bridge sites in the *Moringa oleifera* protein, which results in re-stabilization of unstable particles, as a result of insufficient particle numbers, to form more bridges between particles (Olanrewaju, 2018).

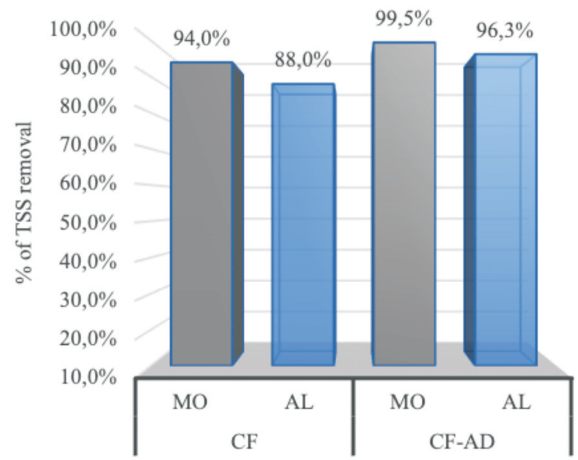

Fig. 4. Effect on TSS removal

In this study, the addition of alum in the coagulation process was able to achieve an efficiency of 88%. This efficiency is due to the addition of alum which has an impact on cations and hydrolyzed products interact with negatively charged colloids so that charge neutralization occurs (Chaouki *et al.*, 2017). Solids that have been neutral then undergo sedimentation to the bottom by gravity under their own gravity which causes a decrease in suspended solids (Raghab *et al.*, 2013; Teh *et al.*, 2016)((Ramprasad *et al.*, 2019) (Gautam and Saini, 2020).

The combination of adsorption in leachate treatment increased the TSS removal efficiency to 99.5% with the addition of *Moringa oleifera* and 96.3%. with the addition of alum. The increase in efficiency is due to the increase in material ingested in the pores of the fly ash adsorbent. TSS removal rates depend on the porosity, media surface, and retention time (Ali *et al.*, 2015). Fly ash as an adsorbent is able to remove TSS from leachate due to its high chemical structure, surface area and porosity. Fly ash pores capture suspended solids and remain until an equilibrium form is reached Ali *et al.*, 2015).

Effect on heavy metal removal

High heavy metal content can have an effect on human health (Shan *et al.*, 2017). Therefore, heavy metals must be treated before being discharged into the environment. The coagulation flocculation with the addition of alum and *Moringa oleifera* aims to remove the heavy metal content in the leachate. The results showed that the addition of alum decreased the pH of raw leachate, whereas the addition of *Moringa oleifera* tended to increase the pH.

The addition of the two coagulants both increased and decreased the pH but still under normal conditions, namely 6-8. pH is considered as two competitive forces, namely 1) between H and the metal hydrolysis products that interact with organic ligands (2) between hydroxide ions and organic anions for interactions with metal hydrolysis products. In this study, pH in the normal range of conditions tends to be alkaline or at high pH values, hydroxide ions compete with organic compounds to adsorb metals so that metals experience joint deposition (Güneº, 2014).

The results showed, *Moringa oleifera seed* was proven to act as a natural coagulant capable of removing heavy metals from leachate. In Figure 5, coagulation flocculation with *Moringa oleifera* is able to remove heavy metals up to 88% while in combination the adsorption efficiency is increased up to 99%. In this study, the removal of oil from the *Moringa oleifera* seed content resulted in the activation of polyelectrolytes and the formation of heavy metal complexes (Meneghel *et al*., 2013). Polyelectrolytes are released after oil extraction because they are insoluble in lipids (Mataka *et al*., 2010; Sajidu *et al*., 2017.).

In the treatment of coagulation flocculation with alum, an efficiency of 78% was obtained. This value increased to 97.4% when combined with adsorption. Alum causes heavy metal to bond to particulate and colloidal materials that are easily absorbed onto the surface of the lump (Hargreaves *et al*., 2018). The increase in removal by adsorption occurs because of the carbon content (Wang *et al*., 2011) and functional oxide groups (SiO_2 and Al_2O_3 that fly ash material is able to absorb heavy metals (Ge *et al*., 2018). At low pH conditions, silica in fly ash is positively charged, whereas at high pH it is negatively charged (Mohan and Gandhimathi, 2009). The zero point charge of silica is generally two. Likewise, Al and Fe also develop surface charges depending on the pH. This shows that silica and alumina in the adsorbent each give a negative charge so that they are able to adsorb metal and metal hydroxide on the adsorbent surface (Sahoo *et al.*, 2013).

The mechanism of fly ash in treating leachate

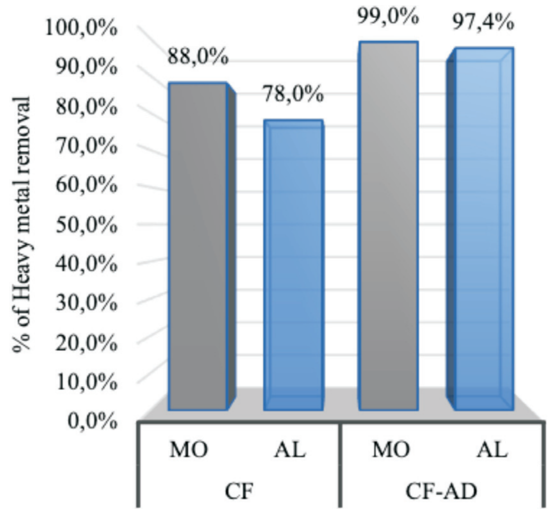

Fig. 5. Effect on heavy metal removal

occurs because of the high adsorption capacity and surface area, as well as the negative charge that accumulates on the surface of the fly ash in a solution that tends to be alkaline. Fly ash can be expected to remove some metal ions from leachate through electrostatic or precipitation-adsorption interactions. The ion exchange capacity, high surface area and unique pore characteristics of fly ash play an important role in processing heavy metals (Ge *et al*., 2018).

CONCLUSION

The results showed that *Moringa oleifera* as a coagulant has a good effectiveness in treating various pollutants contained in leachate, or it can be said to be almost close to alum, even more than TSS and heavy metal. The effectiveness of *Moringa oleifera* compared to alum is that it can remove COD, TSS, and heavy metal respectively by 62%, 94%, and 88% compared to alum at 64%, 88%, and 78%. The combination of adsorption using fly ash increased the effectiveness of *Moringa oleifera* in removing COD, TSS, and heavy metal to 74.2%, 99.5%, and 99.6% respectively compared to 76.6%, 96.3%, and 97.4% alum. Especially for pH with the addition of *Moringa oleifera*, it is necessary to be careful because it tends to be alkaline even though each is in the normal range of 6-8, namely 8.02 with coagulation flocculation and 7.72 with a combination of coagulation and adsorption flocculation. Acid pH of fly ash as adsorbent can reduce pH. Compared with the addition of alum, the pH conditions were closer to normal, namely 7.52 with coagulation flocculation and 7.15 with a combination of coagulation and adsorption flocculation. Although the overall efficiency is quite high, the COD effluent value still does not meet government regulatory standards, therefore it is recommended to combine it with biological treatment in future studies.

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