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Reducing Pollutants in Coal Wastewater with *Moringa oleifera* and Natural Biocoagulants Combination

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ABSTRACT

The wastewater treatment process for middle rank coal employed coagulation-flocculation technology with bio coagulants to efficiently reduce the concentration of contaminants. This method utilized *Moringa oleifera* L. seed powder as the main bio coagulant, in addition to papaya seeds (*Carica papaya* Linn.) and Ambon banana peel waste (*Musa paradisiaca* var.) as secondary bio coagulants, which are combined with the main bio coagulant. The aim of this study was to determine the characteristics and effectiveness of bio coagulant compounds obtained from plants and waste materials. The study objective was also to investigate the effectiveness of combining bio-coagulants derived from *Moringa oleifera* L., *Carica papaya*, and *Musa paradisiaca* var. in treating coal effluent. Biocoagulant is a natural alternative to chemicals like aluminum sulfate (Alum) that are detrimental to the environment. It is present in the ecosystem and serves as a substitute for these hazardous substances. The results revealed that the optimal dosage for combining bio coagulants. The analysis of water pollutants showed a reduction in turbidity and total suspended solid (TSS) by 99.26% and 99.11% respectively. Additionally, there was a decrease in the levels of heavy metals iron (Fe) and manganese (Mn) by 98.71% and 99.88% respectively.

Keywords: coagulation, flocculation, coal wastewater, pollutants, bio coagulant, *Moringa oleifera* L, banana peel waste, papaya seeds.

INTRODUCTION

The mining sector significantly contributes to the overall prosperity of the global economy. Mining resources serve several purposes, including the production of jewellery, ornaments, construction materials, cars, electricity generation, and advanced technological uses (Marimuthu et al., 2021). Significant quantities of water are required for the extraction and refinement of mineral resources, and subsequently, these waters are released back into the environment. The wastewater often above the discharge limits set by the WHO due to its high levels of detrimental organic pollutants, such as flotation chemicals, and inorganic pollutants, such as metal ions (Meißner, 2021).

The discharge of mine effluent into the environment has sparked considerable public apprehension because to its potential to rapidly pollute water sources and inflict harm to aquatic life, terrestrial animals, plants, and human beings (Yaraghi et al., 2020; Thomas et al., 2022). Moreover, the accessibility of potable water is a prevalent issue in numerous global places, especially in arid and semi-arid areas. Wastewater from coal mining operations refers to water that comes from various activities involved in mining coal. These activities include removing the top layer of soil, excavating, dredging, transporting, and storing coal. The water can come from both upstream sources, such as open and closed mines, as well as downstream activities, like loading coal onto barges. The government determines the quality criteria for coal wastewater by referencing SNI 6989 in 2019, APHA 5210 B in 2012, and APHA 5220 D in 2017.

Prior to being released into the environment, the coal mining industry treats the wastewater it generates through a process of water treatment. The objective is to guarantee that the water released adheres to the quality criteria established by the government and does not harm aquatic organisms. Typically, chemical coagulants such as alum $(Al_2(SO_4)_2)$ and iron chloride (FeCl₂) are commonly employed in wastewater treatment operations within the coal mining sector. Chemical coagulants can be employed to filter water prior to its release into the environment (Eri et al., 2018). The benefits of utilizing this coagulant include its cost-effectiveness and ready availability. However, while the use of alum coagulant may be effective in the short term, it is not advisable in the long run due to its negative effects on health, such as Alzheimer's disease, and its environmental impact caused by the disposal of alum residue (Gautama and Saini, 2020; Owodunni and Ismail, 2021). In addition, the presence of high levels of heavy metals, like as manganese, in acid mine drainage poses a risk to human health (Kusdarini et al., 2023).

Moringa seeds (Moringa oleifera L.) and papaya seeds (Carica papaya Linn.) are both efficient bio coagulants utilized in waste treatment. Papaya seeds possess a significant amount of protein, which consists of positively charged proteins capable of binding to negatively charged substances like mud, clay, bacteria, toxins, and so on. The binding process described here causes the aggregation of particles, which then settle and ultimately produce clean water through the processes of adsorption and charge neutralization (Edogbanya, 2013). This elucidates the reason behind the coagulant activities exhibited by papaya seeds. Papaya seed powder can induce the coagulation of suspended particles in water, resulting in their sedimentation at the bottom. The main protein, papain (also known as papaya proteinase), consists of a single, completely connected peptide sequence with 345 amino acid residues (George and Julyn, 2018). In addition, the water has become contaminated with fecal germs due to the introduction of papaya seeds (Kristianto et al., 2018).

The utilization of both bio coagulant and banana peel waste is a substantial type of waste

material. This waste is typically produced during domestic processing and is commonly used as a means of water purification. Previous research has demonstrated that banana peels are 88% effective in reducing turbidity in synthetic wastewater and river water (Mokhtar et al., 2019). In addition, they achieved a 60% reduction in turbidity levels. Furthermore, a study conducted by Maurya and Daverey (2018) revealed that banana peels have a remarkable efficacy of 59.6% in reducing turbidity and TSS in urban wastewater.

The coagulation-flocculation method is frequently used in the wastewater treatment process, as evidenced by previous research. Several recent studies (Camacho et al., 2017; Alam et al., 2020; Katubi et al., 2021) have been done to identify sustainable and ecologically friendly natural coagulants that can be used as alternatives to inorganic and synthetic coagulants in the manufacture of drinking water. Natural coagulants have numerous significant advantages, such as their capacity to biodegrade, their lack of toxicity, their renewable nature (with readily available feedstock), and their cost-effectiveness (both in terms of the coagulant itself and the sludge processing) (Ang and Mohammad, 2020).

Certain natural plants are employed as a coagulant: *Moringa oleifera* L. (Chitra and Muruganandam, 2019; Made et al., 2020; Lelusa et al., 2022), banana peels (Singh et al., 2018; Mokhtar et al., 2019; Musafira et al., 2020), papaya seed (George and Julyn., 2018; Kristianto et al., 2018) and aloe vera (Adugna and Gebresilasie., 2018; Srikanth, 2020). These are some of natural coagulants needed for water and wastewater treatment.

Currently, there is a lack of knowledge regarding the impact of combining bio-coagulants to reduce pollutants in coal wastewater. Specifically, the use of moringa seeds, banana peels, and papaya seeds for medium rank coal wastewater is not well understood. To optimize the results, a comprehensive and methodical investigation was conducted to improve the process by including bio coagulants such as papaya seeds and waste from Ambon banana peels. The objective of this current study is to assess the efficacy of these bio-coagulants in reducing the concentration of contaminants in coal effluent by combining *Moringa oleifera* L, *Carica papaya* Linn., and *Musa paradisiaca* var.

Currently there is only limited study regarding the effectiveness of the mixing bio-coagulants for coal wastewater. The characteristics of medium-rank coal effluent may have significant consequences for both human health and the environment. This study's goal is also to prepare bio-coagulants that can be an alternative to the use of chemical coagulants that are not environmentally friendly. The significance of this study is to examine that the mixing biocoagulants from *Moringa oleifera* L., *Carica papaya* Linn., and *Musa paradisiaca* var. for best options to treat coal wastewater.

The novelty of this study investigates biocoagulants generated from natural sources (Moringa oleifera L., Carica papaya Linn., and Musa paradisiaca var.) as an environmentally acceptable alternative to chemical coagulants in the treatment of coal effluent. This strategy not only solves the critical need for sustainable wastewater treatment solutions, but it also contributes to broader environmental and socioeconomic goals, which are closely aligned with the United Nations Sustainable Development Goals (SDGs). This work directly adds to SDG 6, which aims to guarantee universal access to and sustainable management of water and sanitation. Effective treatment of coal effluent with bio-coagulants improves water quality, reduces contamination, and ultimately protects human health and ecosystems.

MATERIALS AND METHODS

Materials

Materials utilized in this study were medium rank coal wastewater samples, aquadest, sodium periodate reagent (207769-ID), citrate buffer powder (2107669-ID), and FerroVer iron reagent (2105769-ID), moringa seeds (*Moringa oleifera* L.), papaya seeds (*Carica papaya* Linn.), and ambon banana peel waste (*Musa paradisiaca* var.)

Medium-rank coal wastewater was collected at the coal loading port (Jetty) site in Palembang, south Sumatra, Indonesia. It was utilized to treat the wastewater before its release into nearby river systems. The investigation was carried out at the coal wastewater processing facility in Palembang and the laboratory of the Tirta Musi Palembang Regional Drinking Water Company. Bio-coagulant materials, including Moringa Seeds (*Moringa oleifera* L.) were obtained from Kampung Bali, Mariana, Banyuasin, South Sumatera. Papaya seeds (*Carica papaya* Linn.) and Ambon banana peel waste (*Musa paradisiaca* var.) were collected in Ogan Komering Ilir (OKI), south Sumatera, Indonesia. The chemicals utilized include sodium periodate reagent (207769-ID), citrate buffer powder (2107669-ID), and FerroVer Iron Reagent (2105769-ID) were purchased from Sigma Aldrich.

Equipments

Equipment utilized in this research were 50 and 100 mesh sieve, bulbs, EC meters, beaker glass, measuring cup, cuvette, magnetic stirrer, ovens, jar test equipment, tweezers, 10 mL and 1 mL drop pipettes, analytical scales 500 g, and turbidity meter. Plastic sample vials with a capacity of 1 L were used to collect wastewater samples of medium-rank coal. Water quality can be assessed using a range of analytical instruments. The Metrom 632 pH Meter was utilized to evaluate the acidity level (pH), while the Hach 2100Q was employed for measuring turbidity. Furthermore, the Hach DR 900 was utilized to identify the existence of total suspended particles and heavy metals, such as Fe and Mn.

Methodology

The study comprises multiple stages, including the sampling and analysis of middle rank coal, followed by the production of bio-coagulant materials such as Moringa seeds, Papaya seeds, and Ambon Banana peel waste. The bio-coagulants optimum dosages were then determined using a jar test, and the subsequent processes of coagulation, flocculation, and sedimentation were carried out. The water samples were analyzed, and the pollutants' percentage reduction was measured using a spectrophotometer, specifically for turbidity, TSS, and heavy metals Fe and Mn. Figure 1 below illustrated a detailed flow diagram of the process.

Characteristics of medium-rank coal wastewater

Coal wastewater samples were collected at the coal loading port located in Palembang, south Sumatera. The physical properties include TSS, total dissolved solid (TDS), turbidity, and electrical conductivity, whilst the chemical properties comprise the acidity level (pH) and the presence of heavy metals iron (Fe) and manganese (Mn) in wastewater (Mabrouki et al., 2020). The initial



Figure 1. Experiment diagram

volume of the wastewater was diluted by a factor of 10. This is necessary because the instrumentation equipment has a limited reading capacity, which would complicate the analysis procedure if the volume was not diluted.

Table 1 presented the medium-rank coal wastewater characteristics. The attributes of coal wastewater of medium rank may have negative impacts on both human health and the environment. By utilizing the primary bio coagulant, moringa powder, along with the supplementary bio coagulants, papaya seeds and Ambon banana peel waste, it was anticipated that the coagulation-flocculation and sedimentation processes will enhance the water quality metrics. This meets the specified standards for wastewater quality for environmental discharge as outlined in SNI 6989 of 2019, APHA 5210 B of 2012, and APHA 5220 D of 2017.

Bio-coagulant materials preparation

Bio-coagulant resources, such as Moringa Plants, Papaya seeds, and Ambon banana peel waste, sent to a purification procedure to eliminate any impurities that may be present on these materials. Subsequently, the bio coagulant derived from Moringa seeds, papaya seeds, and banana peel waste underwent a thorough rinsing process using clean water. Afterwards, the substance was subjected to a drying process under direct sunlight for a duration of 3–4 days, while maintaining a temperature range of 32–34 °C (Musafira and Dzulkifli, 2020). Prior to the drying

Parameter	Unit	Test result	Environmental quality standard	Methods
Suspended solids	mg/L	3607	199	SNI 6989.3 :2019
рН	_	6.8	6.5–8.5	SNI 6989.11: 2019
Manganese (Mn)	mg/L	2.97	1	APHA 5210 B: 2012
Iron (Fe)	mg/L	3.88	2	APHA 5220 D Ed. 23: 2017

Table 1. Characteristics of medium-rank coal wastewater

process, banana peels were cut into segments that are approximately 2–3 cm long. Subsequently, bio coagulant component performed an equalization procedure by confining it within a specified area and monitoring its mass reached a steady state. The bio-coagulant combination was further crushed using a blender and then strained through a sieve with a size of 100 mesh (0.149 mm).

Determining the optimal dosage using jar test

The main goal of this study was to examine the effectiveness of Moringa seeds as a natural substance that can help remove pollutants from middle rank coal effluent. The dose estimation was dependent on the operating characteristics that are found in the wastewater treatment area. The additional quantity of Moringa coagulant varied from 0.5 g to 1.5 g, with increments of 0.25 g. Each dose was then mixed with 1 liter of wastewater (Nugraha and Sumiyati., 2010). The coagulation-flocculation process consists of initial agitation at a speed of 100 rpm for a duration of 30 seconds, followed by a subsequent slower agitation at 30 rpm for a period of 10 minutes, and ultimately allowing the mixture to settle for 30 minutes. After determining the optimum dose of Moringa bio coagulant, additional bio-coagulants in the form of papaya seeds and Ambon banana peel waste were introduced in different ratios: (1:0), (1:2), (2:1), (1:1), and (0:1). The findings suggested that by optimizing the mixing of bio-coagulants, it was possible to effectively minimize pollutants in medium rank coal effluent. This can be achieved by maintaining the same parameters for the coagulation-flocculation process.

Percentage of pollutants reduction utilizing a spectrophotometer

The objective was to identify the most effective conditions for treating medium rank coal wastewater using Moringa bio-coagulant. This involved adjusting the parameter values or removal percentages for each concentration of Moringa and incorporating additional bio coagulants like papaya seeds and Ambon banana peel waste. The water quality parameter values were measured using the Hach DR 2001Q type Spectrophotometer to determine the TSS values, and the Hach DR 900 type Spectrophotometer to analyze the concentrations of the heavy metals Fe and Mn.

These values were then compared to the medium rank coal wastewater quality standards specified in the SNI 6989 standard. The APHA 5210 B standard in 2012, and the APHA 5220 D standard in 2017. The percentage of removal was calculated by comparing the change in each parameter value to the control treatment. To enhance the extent of parameter reduction in wastewater quality with the use of Moringa bio-coagulant, the water was examined both prior to and following the addition of the bio-coagulant. The allowed percentage was determined using the formula provided by Linggawati (Linggawati, 2002).

$$R = \frac{(A-B)}{A} x \ 100\% \tag{1}$$

where: R – removal percentage (%); A – results of water quality analysis before treatment (control); B – results of water quality analysis after treatment.

This removal percentage formula can be used for various parameters such as turbidity, TSS, heavy metals Fe and Mn.

RESULT AND DISCUSSION

Bio-coagulants production

The main bio coagulant utilized was derived from Moringa seeds, sourced from two distinct origins: commercial Moringa and local Moringa (Figure 2 displays the raw materials). The weight of the commercial Moringa seeds was 60.70 g, whereas the weight of the local Moringa seeds is 13.99 g. The amount of bio coagulant utilized consisted of 51.81 g of papaya seeds and 72.56 g of Ambon banana peel waste. After being dried in the sun for a duration of 3–4 days, the major bio coagulants, specifically the commercial and local moringa bio coagulants, were weighed and found to be 60.63 g and 13.49 g respectively. Furthermore, the bio-coagulants used for support, specifically the papaya seeds and Ambon banana peel waste, weighed 8.39 g and 7.47 g respectively. Afterwards, the bio-coagulation material underwent an equilibration procedure at a room temperature of 27-28 °C until its weight stabilized. Subsequently, it was crushed utilizing a blender. The weight of each bio-coagulant material was measured as follows: the commercial moringa, local moringa, papaya seeds, and banana peel waste had weights of 60.68 g, 13.68 g, 8.53 g, and 7.61 g, respectively. Figure 2 illustrated the procedure for preparing the bio-coagulant materials.

The effect of the dosage moringa seeds (*Moringa oleifera* L.) on the reduction of total suspended solids

An experiment was conducted to evaluate the efficacy and efficiency of Moringa seeds as the primary bio coagulant in decreasing pollutants in medium-rank coal wastewater. This was done before the addition of papaya seeds and Ambon banana peel waste as secondary bio coagulants. This experiment involved using five measuring glasses, each with a capacity of 1000 mL, to prepare samples with different doses of Moringa seeds. The doses used were 0.5 g, 0.75 g, 1 g, 1.25 g, and 1.5 g. The purpose of this experiment was to compare the quality of commercial Moringa seeds with local Moringa seeds in terms of their ability to reduce pollutants in wastewater.

According to Figure 3, the test findings demonstrated that using 1 gram of the commercial moringa coagulant per liter of water led in a TSS value of 29.3 mg/L, with a percentage of 99.19%. In contrast, the TSS concentration of natural moringa was measured at 21 mg/L, with a removal rate of 99.42%. To effectively remove high levels of TSS from water, which includes both organic and inorganic pollutants, it is crucial to determine the exact dosage or concentration of a coagulant. This will ensure the best settling of the colloidal particles that contribute to TSS.

Moringa seeds contain polyelectrolyte macromolecular proteins with positive charges. When these proteins break down, they attract negatively charged particles, leading to an increase in TSS. When using a dose of 1.25 g/L of commercial moringa coagulant and a dose of 1.5 g/L of natural moringa, the TSS decreased. This decline occurred because the concentration of the coagulant



a) Moringa Seed (commercial) b) Moringa Seed (local)



Figure 2. Bio-coagulant materials preparation



Figure 3. Effect of Moringa seed dosage on reducing TSS

surpassed the optimal level, causing deflocculation. As a result, the TSS value increased again (Hak et al., 2018). In addition, despite the use of a 100-grit screen for filtration, the coagulant particles exhibited non-uniform size, resulting in their retention and subsequent re-coagulation (restabilization). The seed powder contains a variety of proteins that are water soluble cations.

According to reports, the water-soluble dimeric cationic proteins found in the seeds of Moringa oleifera L. were what give the plant its coagulant properties. This implies that the water solution would become basic because of the basic amino acids included in the proteins of Moringa oleifera L. seeds accepting a proton from the water and releasing a hydroxyl group (Hendrawati et al., 2016). Proteins function as natural polyelectrolytes, possibly through a combination of adsorption, charge neutralization, and particle bridging of destabilized particles (Camacho et al., 2017). Adsorption and charge neutralization are the primary activities associated with Moringa seeds, although it is challenging to determine the specific mechanism at work due to their simultaneous occurrence (Hendrawati et al., 2016). When moringa seeds were introduced into turbid water, it was expected that the negatively charged particles, such as dust, bacteria, clay, and others, will adhere to the protein in the seeds, resulting in the formation of bigger flocs that can settle down.

Although this study focused primarily on the effects of coagulant dose, flocculant dose, and agitation time, it is important to note that the treatment's success was dependent on other parameters such as agitation speed. It was critical to understand the role of the applied velocity gradient in the coagulation flocculation process. The velocity gradient was an important factor in determining the likelihood of particles colliding during coagulation and flocculation. This contact is critical for floc development and proliferation, which has a direct impact on the overall efficacy of the treatment process. Excessive velocity can cause mechanical shear pressures that destabilize flocs and reduce their ability to agglomerate suspended particles (Jebun et al., 2018; Adesina et al., 2019; Djeffal et al., 2021).

Effect of dosage Moringa seeds (*Moringa oleifera* L.) in reducing iron and manganese metals

Heavy metals are hazardous contaminants that could build up in the environment and combine with both organic and inorganic substances in aquatic environments, resulting in the formation of intricate compounds. Exceeding tolerance limitations of heavy metals can harm human physiological and biological systems. Heavy metals are introduced into the tissues of organisms by various pathways, including the respiratory system, the digestive system, and absorption through the skin (Anthony and Godfrey., 2014). The heavy metal content tested in this study included Fe and Mn. These two elements, classified as heavy metals, form part of the chemical composition of middle rank coal effluent.

The presence of heavy metals in medium rank coal wastewater primarily originates from the coal rock's formation layers, which collected in the rock's core layer. The initial concentrations of the heavy elements Fe and Mn in middle rank coal effluent were 3.88 mg/L and 2.97 mg/L, respectively. Fe metal analysis was conducted for each dosage of both commercial and natural Moringa coagulant.

Figure 4 depicts the results of the analysis of the heavy metal Fe in effluent from medium rank coal. The wastewater underwent treatment by adding Moringa coagulant, and the analysis considered various dosages and particle sizes. The content of the heavy metal Fe was significantly reduced by 99.10% for commercial moringa and 97.16% for natural moringa when a dose of 1 g/L was used, considering other factors that may have contributed to the reduction. The obtained findings meet the quality criteria for mediumgrade coal effluent, enabling its safe discharge into the environment. More precisely, the outcomes meet the criteria of 2 mg/L as stipulated by APHA 5220 D of 2017.

Figure 3 illustrates that when the coagulant dose of 1.25 g/L is applied for both commercial and natural moringa, which exceeds the recommended amount, the efficiency of eliminating the heavy metal Fe would diminish. The phenomenon happens when the coagulant has achieved the optimal dosage, resulting in an excess or insufficient amount of coagulant being saturated and forming particles that are challenging to settle (Rani, 2010). Moringa seeds contain amino acids with a low molecular weight. These amino acids form a biologically active group of molecules that can bind to metals even at low concentrations, facilitating the absorption of metal ions. In addition, these amino acids, which are composed of proteins, have pH-dependent characteristics that

create an environment with a negative charge. This negative charge is crucial in facilitating the binding of heavy metals (Costa et al., 1997; Muyibi et al., 2002). Therefore, the inclusion of a higher dosage did not improve the effectiveness of a coagulant.

The analysis of Mn metal was conducted using both commercial and natural moringa at an optimal dose of 1 g/L. Table 2 displays the reduction in manganese content resulting from the application of varying quantities of commercially available Moringa seeds and naturally occurring Moringa seeds. The concentration of natural moringa decreased by 0.052 mg/L, while the concentration of commercial moringa reduced by 0.050 mg/L. The objective of this study was to quantify the extent of Mn metal reduction using Moringa coagulant. Subsequently, the coagulant was combined with Ambon banana peel and papaya seed coagulant to assess the impact of their addition. Moringa seeds, scientifically known as Moringa oleifera L. are frequently utilized as biocoagulants due to their abundant protein content. These seeds can effectively serve as biocoagulants in waste treatment processes owing to the presence of positively charged proteins (Edogbanya, 2013). These proteins can bind with negatively charged particles such as mud, clay, bacteria, and toxins, resulting in the formation of clumps that settle down and yield purified water through the processes of adsorption and charge neutralization.

 Table 2. Effect of Moringa seed dosage on reducing manganese (Mn)

Natural Moringa seed	Commercial Moringa seed	
(mg/L)	(mg/L)	
0.052	0.050	



Evaluation of the efficacy of combining papaya seed bio-coagulant with Ambon banana peel waste

Combination of Moringa seeds and Papaya seeds for bio-coagulant I

This study utilized two natural coagulants, namely papaya seeds (*Carica papaya* Linn.) and Ambon banana peel waste (*Musa paradisiaca* var.), as mixing components with *Moringa oleifera* L. seeds. The concentration of Moringa was 1 g/L. The mixing ratio for combining bio coagulant I (made from Moringa seeds and Papaya seeds) and bio coagulant II (made from Moringa seeds and Ambon banana peel) was in a ratio of 1:0, 1:1, 1:2, 2:1, and 0:1. Afterwards, the findings from the investigation of test parameters in medium rank coal waste water will be contrasted to determine the most favorable conditions for combining the two bio coagulant substances.

Table 3 and 4 demonstrated that when papaya seeds (*Carica papaya*) were added in a dose ratio of 0.25 g papaya: 0.75 g moringa, or a ratio of 1:2, under ideal conditions, they successfully decreased pollutant levels, particularly heavy metals Fe. When 0.25 g of papaya was mixed with commercial coagulants, the resultant levels decreased to 0.05 mg/L, whereas the natural coagulants had a higher iron level of around 0.31 mg/L. The TSS concentration was 32 mg/L when

using a combination of commercial coagulants, and roughly 51 mg/L when using a mixture of natural moringa.

Ningsih (2020) states that TSS deficiency is caused by the presence of protein in the water, and the consumption of papaya seeds yields beneficial outcomes in a significant proportion of samples. The pH data did not indicate any significant variation between the different dosages of both the commercial and natural coagulants. The size of the coagulant particles directly affects this value. If the size is not optimized, it might result in inadequate waste processing outcomes, hence reducing the effectiveness and efficiency of the coagulation-flocculation method. Conversely, if the size of the coagulant particles is very large, it will cause the scattered particles to disperse, thus disrupting the particle deposition process (Ngadi and Aida, 2013).

Papaya seeds exhibited significant efficacy in reducing the content of heavy metals, such as Mn, in middle rank coal effluent. Using 1 gram of moringa, the parameter value for Mn metal was 0.05 mg/L for commercial moringa and 0.052 mg/L for natural moringa. With the inclusion of papaya seeds, there was a significant decrease in the concentration of Mn metal. More precisely, there was a reduction of 0.004 mg/L (corresponding to a 93% decrease in commercial moringa) and a reduction of 0.008 mg/L (equivalent to an 85.48% decrease in natural moringa) when the bio

<u>_</u>	5	0	
Coogulant doorgo (g)	Test parameter		
Coaguiant dosage (g)	рН	TSS (mg/L)	Fe total (mg/L)
1 g (papaya seed)	6.85	132.00	0.27
0.75 g (papaya seed)	6.84	42.00	0.15
0.5 g (papaya seed)	6.86	17.50	0.06
0.25 g (papaya seed)	6.80	32.00	0.05
1 g (moringa)	6.98	29.30	0.04

Table 3. Results of analysis of adding papaya seeds to commercial Moringa

Note: Mn analysis at the optimum dose of 0.25 g papaya is 0.004 mg/L

Table 4. Results of analysis of adding papaya seeds to natural Moringa

	Test parameter			
Coaguiant dosage (g)	рН	TSS (mg/L)	Fe Total (mg/L)	
1 g (papaya seed)	6.85	132.00	0.27	
0.75 g (papaya seed)	6.84	219.50	0.38	
0.5 g (papaya seed)	6.88	60.50	0.20	
0.25 g (papaya seed)	6.84	51.50	0.31	
1 g (moringa)	6.62	21.00	0.11	

Note: Mn analysis at the optimum dose of 0.25 g papaya is 0.008 mg/L

coagulant was mixed under optimal conditions (with a ratio of 1 part papaya to 2 parts moringa).

The inclusion of papaya seeds (*Carica papaya* Linn.) will enhance the ability to draw metals into the floc. The decrease in metal levels is ascribed to the interaction between the cationic protein present in papaya seed powder and the negatively charged component that binds the metal ions. The binding process enhances the formation of solid metal ions (Ali et al., 2019).

Combination of Moringa seeds with banana peel for bio-coagulant II

In this study, an additional natural coagulant, Ambon banana peel waste (Musa paradisiaca var.), was included as a mixing material together with Moringa oleifera L. seeds. The bio coagulants were mixed at the ideal dosage of 1 g/L using Moringa seeds. The mixing ratios for bio coagulant II include 1:0, 1:1, 1:2, 2:1, and 0:1. Subsequently, the findings from the measurement of test parameters for medium rank coal wastewater was contrasted with different coagulant materials that exhibit superior efficacy in reducing contaminants in medium rank coal wastewater. The efficacy of integrating Ambon banana peel waste in reducing pollutant levels, particularly in relation to TSS and the heavy metal Fe, was illustrated by Tables 5 and 6. When combining papaya seeds, adding a dose ratio of 0.25 g of banana seeds to 0.75 g of moringa, or a ratio of 1:2, significantly reduced the level of pollutants. When 0.25 g of papaya were combined with commercial coagulants, the TSS readings were found to be 14.33 mg/L. On the other hand, when natural mixed coagulants were used, the TSS values were 52.80 mg/L. The utilization of Ambon banana peel leads to a drop in TSS content, indicating that the banana peel can adsorb dissolved particles in wastewater, hence causing a reduction. The intensity of the contact increases with the duration of the circulation. This aligns with the findings of Hanifah and Hadisoebroto (2020) study, which revealed that banana peel is rich in minerals, vitamins, proteins, carbohydrates, and lipids. Protein is a crucial element in the process of coagulation. This is the process that leads to the purification of wastewater and the reduction of TSS.

The combination of 0.75 g of commercial moringa and 0.25 g of banana also led to a decrease in Mangan content by 0.021 mg/L. Although natural moringa was added, no significant change was found, especially in the reduction of the pollutant Mn, which had a concentration of 0.028 mg. In their study, Budiman et al. (2018) discovered that the peel of Kepok banana (Musa acuminate) can absorb metal ions, making it a potential biomaterial for this purpose. Cellulose is a constituent of Ambon banana peel. The cellulose in Ambon banana peels enables them to adsorb metal ions. Cations will be drawn to hydroxyl groups that possess a surplus of electrons. Disposing of Ambon banana peels can efficiently reduce the concentration of Fe²⁺ ions in aqueous solutions. Ambon banana peels possess the capacity to efficiently absorb manganese ions found in well water.

The optimal removal of Fe metal was achieved by combining 0.25 g of banana peel with commercial moringa at a concentration of 0.13 mg/L, resulting in a removal efficiency of 96.73%. The utilization of natural moringa at a concentration of 0.17 mg/L resulted in a removal effectiveness of 95.53%. The low efficiency is due to the use of powdered natural coagulant obtained from discarded Ambon banana peels. Furthermore, the effect of a low stirring velocity of 30 revolutions per minute (rpm) should be considered. Consequently, reducing the stirring or flocculation speed leads to a more significant elimination of Fe metal in medium rank coal effluent. Angraini et al. (2016) demonstrated that when stirring speeds are low, the attractive forces between colloid particles become stronger and overcome the repulsive forces, resulting in the formation of bigger flocs.

Table 5. Results of analysis of the addition of ambon banana peel to commercial Moringa

Coogulant docogo (g)	Test parameter		
Coaguiant dosage (g)	рН	TSS (mg/L)	Fe total (mg/L)
1 g (banana peel)	6.44	140.00	0.47
0.75 g (banana peel)	6.49	81.67	0.25
0.5 g (banana peel)	6.46	45.33	0.14
0.25 g (banana peel)	6.43	14.33	0.13
1 g (moringa)	6.98	29.30	0.04

Note: Mn analysis at the optimum dose of 0.25 g of banana is 0.021 mg/L

	Test parameter		
Coaguiant dosage (g)	рН	TSS (mg/L)	Fe total (mg/L)
1 g (banana peel)	6.44	354.00	0.47
0.75 g (banana peel)	6.41	203.00	0.44
0.5 g (banana peel)	6.41	124.00	0.30
0.25 g (banana peel)	6.46	52.80	0.17
1 g (moringa)	6.62	21.00	0.11

Table 6. Results of analysis of the addition of Ambon banana peel to natural Moringa

Note: Mn analysis at the optimum dose of 0.25 g of banana is 0.028 mg/L

CONCLUSIONS

This work stands out in that it takes an innovative approach to wastewater treatment by using bio-coagulants derived from natural sources, providing a sustainable alternative to chemical coagulants. This research tackles immediate environmental concerns while also contributing to broader global sustainability efforts by aligning with key Sustainable Development Goals such as clean water and sanitation.

The effectiveness of bio-coagulants in treating medium rank coal wastewater was significantly affected by the dosage and particle size. Utilizing the ideal dosage and particle size ensured that the coagulation-flocculation process operates at its optimal level, leading to a reduction in the physical and chemical properties of middle rank coal effluent. The study determined that the ideal dosage for the primary bio coagulant derived from *Moringa oleifera* L. seeds were 1 gram per liter.

The analysis of medium rank coal wastewater, treated with a bio coagulant made from papaya seeds (*Carica papaya* Linn.) and commercial moringa, yielded significant results. The addition of the bio coagulant resulted in TSS showed a reduction of 99.11%. Furthermore, the heavy metal content of Fe and Mn decreased by 98.71% and 99.88% respectively. The results suggest that the addition of the papaya seed bio-coagulant mixture significantly improved the quality of wastewater, bringing it very close to the minimum quality standards set for coal processing/washing wastewater in SNI 6989 of 2019, APHA 5210 B of 2012, and APHA 5220 D of 2017.

The analysis of medium rank coal wastewater, treated with a bio coagulant derived from Ambon banana peel waste (*Musa paradisiaca* var.), yielded the following results: TSS of 99.60%, and a significant reduction in the concentrations of the heavy metals Fe and Mn by 96.73% and 99.30% respectively. The results demonstrated that the addition of the bio-coagulant mixture derived from wasted banana peels of the Ambon banana variety improves the quality of wastewater.

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REFERENCES

- Adesina, O.A., Abdulkareem, F., Yusuff, A.S., Lala, M., and Okewale, A., 2019. Response surface methodology approach to optimization of process parameter for coagulation process of surface water using *Moringa Oleifera* seed. South Afr. J. Chem. Eng. 28, 46–51. https://doi. org/10.1016/j.sajce.2019.02.002
- Adugna, A.T., and Gebresilasie, N.M., 2018. Aloe steudneri gel as natural flocculant for textile wastewater treatment. Water Pract. Technol. 13(3), 495– 504. https://doi.org/10.2166/WPT.2018.062.
- Alam, M.W., Pandey, P., Khan, F., Souayeh, B., and Farhan, M., 2020. Study to investigate the potential of combined extract of leaves and seeds of *Moringa Oleifera* in groundwater purification. Int. J. Environ. Res. Public Health 17(20), 1–13. https://doi. org/10.3390/ijerph17207468
- Ali, M., Mustafa, A., and Saleem, M., 2019. Comparative study between indigenous natural coagulants and alum for microalgae harvesting. Arabian Journal for Science and Engineering, 44(7), 6453–6463.
- Ang, W.L., Mohammad, A.W., 2020. State of the art and sustainability of natural coagulants in water and wastewater treatment. J. Clean. Prod. 262, 121267 https:// doi.org/10.1016/j. jclepro.2020.121267. Elsevier Ltd.
- 6. Angraini, S., Pinem, J.R., Saputra, E. 2016. The

effect of stirring speed and pumping pressure on the combination of coagulation processes and ultrafiltration membranes in rubber industry liquid waste processing. Bachelor's Thesis, Faculty of Engineering, Riau University Pekanbaru. (in Indonesia).

- 7. Anthony, F. and Godfrey, K., 2014. Simultaneous adsorption of Ni(II) and Mn(II) Ions from aqueous solution into a nigerian kaolinite clay. Integrative Medicine Research, 3(2), 129–141.
- Budiman, H., and Hasria. 2018. Kepok banana peel waste (*Musa Acuminate*) as a biofilter for Iron (Fe) and Lime (CaCO₃). Promotive: Journal of Public Health, 8(2), 152–158. (in Indonesia).
- Camacho, F.P., Sousa, V.S., Bergamasco, R., Teixeira, M.R. 2017. The use of *Moringa oleifera* as a natural coagulant in surface water treatment. Chem. Eng. J. 313, 226–237, https://doi.org/10.1016/j.cej.2016.12.031
- Chitra, D., and Muruganandam, L. 2019. Performance of natural coagulants on greywater treatment. Recent Innov. Chem. Eng. Former. Recent Pat. Chem. Eng. 13(1), 81–92. https://doi.org/10.2 174/2405520412666190911142553
- Costa, G., Michant, J.C., Guckert, G. 1997. Amino acids exuded form cadmium concentrations. Journal of Plant Nutrition. 20, 883–900.
- 12. Djeffal, K., Bouranene, S., Fievet, P., Déon, S., Gheid, A., 2021. Treatment of controlled discharge leachate by coagulation flocculation: Influence of operational conditions. Sep. Sci. Technol. 56, 168–183. https://doi. org/10.1080/01496395.2019. 1708114
- Edogbanya. 2013. A Review on the use of plants' seeds as biocoagulants in the purification of water. Continental J. Biological Sciences, 15(3–2), 26–32.
- Eri, I.R., Hadi, W., Slamet, A. 2018. Clarification of pharmaceutical wastewater with *Moringa Oleifera*: Optimization through response surface methodology. Journal of Ecological Engineering. 19(3), 126–134. https://doi.org/10.12911/22998993/86148
- Gautama, S., and Saini, G. 2020. Use of natural coagulant for industrial wastewater treatment. Glob. J. Environ. Sci. Manag. 6, 553–578.
- George, D., and Julyn, A.C. 2018. Coagulation Performance Evaluation of Papaya Seed For Purification Of River Water 50–66.
- Hak A., Kurniasih, Y., Merdana, I.M. 2019. Effectiveness of using *Moringa Oleifera* seed powder as a coagulant to reduce TDS and TSS in laundry waste. Journal of Chemical Education. 6(2), 2338–6487. (in Indonesian).
- 18. Hanifah, H.N. and Hadisoebroto. 2020. Effectiveness of biocoagulant of purebred chicken egg shells and kepok banana peel (*Musa Balbisiana ABB*) in reducing turbidity, TDS, and TSS from Liquid Waste from the Pharmaceutical Industry. Al-Kimiya, 7(1),

47-54. (in Indonesian).

- Hendrawati, Yuliastri, I.R., Nurhasni, Rohaeti, E., Effendi, H., Darusman, L.K., 2016. The Use of *Moringa Oleifera* Seed Powder As Coagulant to Improve The Quality of Wastewater and Ground Water, IOP Conf. Ser.: Earth Environ. Sci. 31, 012033, https:// doi.org/10.1088/1755-1315/31/1/012033
- 20. Jebun, N., Al-Mamun, A., Alam, Md.Z., Raus, A.R., 2018. Optimization of Flocculation Process for A New Myco-Coagulant to Reduce Water Turbidity. In: Yacob, N.A., Mohd Noor, N.A., Mohd Yunus, N.Y., Lob Yussof, R., Zakaria, S.A.K.Y. (Eds.), Regional Conference on Science, Technology and Social Sciences (RCSTSS 2016). Springer, Singapore, 271–281. https://doi. org/10.1007/978-981-13-0074-5_25
- 21. Katubi, K.M., Amari, A., Harharah, H.N., Eldirderi., M.M., Tahoon, M.A., Rebah, D.B., 2021. Aloe vera as promising material for water treatment: A review. Processes. MDPI AG. https://doi. org/10.3390/pr9050782
- Kristianto, H., Kurniawan, M.A., Soetedjo, J.N.M., 2018. Utilization of Papaya Seeds As Natural Coagulation For Synthetic Textile Coloring Agent Wastewater Treatment. I.J.A.S.E.I.T. 2071–2077.
- 23. Kusdarini, E., Sania, P.R., Budianto, A., 2023. Adsorption of iron and manganese ions from mine acid water using manganese green sand in batch process. 24(12), 158–166. https://doi. org/10.12911/22998993/173007
- 24. Lelusa, G.A. 2022. Water treatment using natural coagulant and electrocoagulation process: А comparison study. Int. J. Anal. Chem. https://doi.org/ 10.1155/2022/4640927
- 25. Linggawati, A., Muhdarina, and Sianturi, H., 2002. Effectiveness of starch-phosphate and aluminum sulfate as flocculants and coagulants. Indonesian Nature Journal, 4(1). (in Indonesian).
- 26. Mabrouki, J., Moufti, A., Bencheikh, I., Azoulay, K., El Hamdouni, Y., El Hajjaji, S. 2020. Optimization of the coagulant flocculation process for treatment of leachate of the controlled discharge of the City Mohammedia (Morocco). In: Ezziyyani, M. (Ed.), Advanced Intelligent Systems for Sustainable Development (AI2SD'2019), Lecture Notes in Electrical Engineering. Springer International Publishing, Cham, 200–212. https://doi. org/10.1007/978-3-030-36475-5_19
- 27. Made, I.A.U, and Merdana, I.M., 2020. Giving Moringa Seed Powder (*Moringa Oleifera*) Reduces the acidity level of pig farm liquid waste. Udayana University: Bali. (in Indonesia).
- Marimuthu, R., Sankaranarayanan, B., Ali, S.M., de S. Jabbour, A.B.L., Karuppiah, K., 2021. Assessment of key

socio-economic and environmental challenges in the mining industry: Implications for resource policies in emerging economies, Sustain. Prod. Consum. 27, 814–830, https://doi.org/10.1016/J. SPC.2021.02.005

- 29. Maurya, S., and Daverey, A. 2018. Evaluation of plant-based natural coagulants for municipal wastewater treatment. Journal Biotech School of Environment and Natural Resources, Doon University, Dehradun, Uttarakhan, India.
- 30. Meißner, S. 2021. The impact of metal mining on global water stress and regional carrying capacities—a gis-based water impact assessment, Resources 10, 120, https://doi.org/10.3390/ RESOURCES10120120/S1
- Mokhtar, N.M., M. Priyatharishini, R.A. Kristanti., 2019. Study on the effectiveness of banana peel coagulant in turbidity reduction of synthetic wastewater, Int. J. Eng. Technol. Sci. 6
- 32. Musafira and Dzulkifli, 2020. Absorption of mercury metal ions using activated charcoal from Kepok banana peel waste (*Musa paradisiaca formatypica*). Covalent: Journal of Chemical Research, 6(1), 39–44. (in Indonesia).
- Muyibi, S., Noor., M.J.M.M., Leong, T.K., Loon, L.H., 2002. Effects of oil extraction from *Moringa Oleifera* seeds on coagulation of turbid water. International Journal of Environmental Studies. 59, 243–254.
- 34. Ngadi, N., and Aida, N., 2013. Treatment of textile wastewater using biodegradable flocculants of chitosan and extracted pandan leaves. Technology Journal, 1, 1–7.
- 35. Ningsih, N.R., 2020. Effectiveness of melon seeds (*Cucumis melo* L.) and papaya seeds (*Carica papaya* L.) as natural coagulants to reduce

pollutant parameters of tofu industrial wastewater. Thesis. Environmental Engineering, Faculty of Science and Technology, UIN Sunan Ampel Surabaya. (in Indonesian).

- 36. Nugraha, S. and Sumiyati., 2010. Wastewater treatment for coal mining activities using biocoagulants: Study of reducing TSS, Total Fe and total Mn levels using Moringa seeds (*Moringa Oleifera*). Diponegoro University: Semarang. (in Indonesian)
- Owodunni, A.A., Ismail, S., 2021. Revolutionary technique for sustainable plant-base green coagulants in industrial wastewater treatment – A review. J. Water Process Eng. 42, 102096.
- 38. Rani, I.Y., 2010. Use of Moringa oleifera Seed Powder as a Coagulant and Flocculant in Improving the Quality of Wastewater and Groundwater. UIN Syarif Hidayatullah: Jakarta. (in Indonesian)
- Singh, S., Parveen, N., Gupta, H. 2018. Adsorptive decontamination of rhodamine-b from water using banana peel powder: A biosorbent. Environ. Technol. Innov. 12, 189–195. https://doi.org/10.1016/j.eti.2018.09.001
- 40. Srikanth, K., 2020. Study on replacement of alum with aloe vera gel in turbidity removal. Int. J. Eng. Res. Appl. 10(8), 17–19. https://doi. org/10.9790/9622-1008031719
- 41. Thomas, G., Sheridan, C., Holm, P.E., 2022. A Critical review of phytoremediation for acid mine drainage-impacted environments, Sci. Total Environ. 811, 152230, https://doi.org/10.1016/J. SCITOTENV.2021.152230
- 42. Yaraghi, N., Ronkanen, A.K., Haghighi A.T., Aminikhah, M., Kujala, K., Kløve, B., 2020. Impacts of gold mine effluent on water quality in a pristine sub-arctic river. J. Hydrol. 589, 125170, https://doi.org/10.1016/J.JHYDROL.2020.125170