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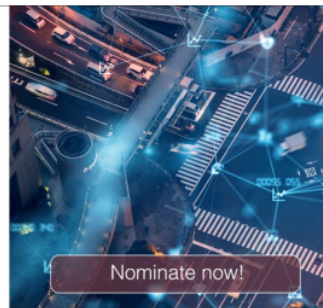
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The feasibility of combined coagulation flocculation and constructed wetland as green technology for sustainable leachate treatment

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Abstract. This study aims to determine the feasibility of coagulation flocculation processing using Moringa oleifera seed powder (MOSP) and vertical sub surface flow constructed wetlands (VSSFCW) to process leachate landfill (LL). The method used is to do a jar test to determine the optimum conditions and then proceed with VSSFCW processing. In the coagulation flocculation process, the optimum conditions were obtained with a dose of 175 mg/L, pH 6.5 and a speed of rapid mixing of 100 rpm for 10 minutes. This optimum condition is able to reach the percentage of COD, turbidity and Mn removal of 35.27%, 69.48%, and 94.83%, respectively. However, the effluent value is still above the Minister of Environment regulation standard No. 5/2014 regarding the standard quality of wastewater for the parameters of COD and Mn that is equal to 584.19 mg/L for COD and 6.98 mg/L for Mn, while for turbidity meets the standard of 69.83 mg/L. Further processing with VSSFCWs can improve the quality of LL effluent resulting from coagulation flocculation 66.36 mg/L; 4.97 mg/L for turbidity and 0.12 mg/L for Mn or with efficiencies of 88.64%, 92.88% and 98.22%. These results were obtained at the optimum HRT conditions for 10 days.

1. Introduction

The increase in population and changes in lifestyle have led to a significant increase in municipal solid waste (MSW) production. The majority of MSW worldwide or almost 95% are disposed of using sanitary landfills [1]. However, the waste processing method produces leachate which contains various types of pollutants that are harmful to human health, flora & fauna and ecosystems [2]. Leachate is a liquid formed from rain water that falls on landfills containing chemical oxygen demand (COD), biological oxygen demand (BOD), ammonia, heavy metals, organic salts, phenols, total dissolved solid (TDS), alkali, hardness, chloride, sulfide, and phosphorus [3-5].

The type of leachate treatment depends on its biodegradability which is known from the value of the BOD/COD ratio. Intermediate leachate is in the range of the BOD/COD ratio of 0.05-0.2, while the stable leachate has a BOD/COD ratio value <0.1 and a BOD/COD ratio value > 0.1 for young leachate [6]. The first technology choice for young and intermediate leachate processing is always on biological processing such as constructed wetland [7], aerated lagoon and rotating biological contactor [8] because of its easy and inexpensive operation [9]. But in mature leachate (BOD₅/COD <0.1), processing must use physical chemistry such as flocculation-coagulation, chemical precipitation or advanced oxidation process to be discharged directly into the environment or to increase its biodegradability so that it can



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be further processed in biological processing. The majority of wastewater treatment plants today are heavy installations of non-renewable materials such as concrete that consume large amounts of energy and chemicals and require high operational and maintenance costs [10]. Therefore, it is necessary to have an infrastructure that adopts a modern outlook and emphasizes the ecological value in wastewater treatment so that it supports sustainable development called green technology.

Green technology in wastewater treatment aims to conserve water usage, prevent contamination, as well as protect ecosystems [11]. The need for new infrastructure that meets processing and sanitation requirements and can create green spaces in urban environments is inevitable. Constructed wetland is an ideal choice that can provide added value to create green ecosystems while maintaining water circulation in urban areas [12]. Constructed wetland is a man-made system that is designed, built and operated to mimic natural wetlands [13]. The system is an economical alternative to wastewater treatment because it uses local resources and is energy efficient [14]. The treatment process in this system utilizes plants and microorganisms so that they are able to process various volumes of wastewater and various contaminants [15,16]. Constructed wetland is able to eliminate various types of pollutants while improving water quality through a series of physical, chemical and biological processes [17]. The weakness of constructed wetland is the need for a large area. The use of an aerobic pre-treatment before constructed wetland is one way to reduce the area required as well as removal of organic matter content [18] Removal of organic material in this pretreatment also serves to prevent clogging, thus prolonging the lifetime of constructed wetland [19].

One type of pretreatment is a combination of physical and chemical processes [20]. Coagulation flocculation is the oldest type of leachate treatment for both leachates that have been stable [21] and intermediate [22]. In this step, chemicals are added to form the final product which is insoluble. In addition this process aims to eliminate various leachate parameters including organic materials which cannot be biodegradable through ionic mechanisms [4]. Chemicals added to the coagulation flocculation process are called coagulants. In general, the coagulant used is synthetic coagulant such as aluminum polychloride, aluminum sulfate and iron chloride. Aluminum polychloride is the most widely used type at present [23].

The disadvantage of using synthetic coagulant is the residual aluminum content in effluent treatment which can trigger Alzheimer's disease [24]. Therefore, natural coagulant is an attractive alternative because of its abundant availability, low cost, non-toxicity, and multifunctionality [25]. In many countries there are many plants and their derivatives that contain natural biopolymers used as coagulants including *Moringa oleifera* (MO). MO is widely used in drinking water and wastewater treatment due to the presence of water-soluble cationic peptides with a molecular weight of 6-16 kDa which can reduce turbidity up to 92-99% [26]. The effectiveness achieved is almost similar to alum but produces less mud and is not dangerous because all MO by products are organic, non-toxic and biodegradable [27]. This study aims to determine the effectiveness of a combination of constructed wetland treatment with flocculation-coagulation pretreatment for leachate landfill treatment.

2. Methodology

2.1. Material

2.1.1. *Leachate*. Leachate landfill is taken as much as 40 liters using grab sampling method and stored at 4 ° C according to methods for the examination [28]. Previously used for the experiment the sample was removed from cold storage and left at room temperature first. Samples of raw leachate were analyzed for temperature, pH, Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), Turbidity using the standard method and heavy metals namely manganese (Mn) using an Inductively Coupled Plasma Mass Spectrometer (Perkin Elmer-Elan 6000, 1999) to determine the efficiency of treatment. The preliminary characteristics of raw leachate can be seen in Table 1.

Table 1. Characteristics of raw leachate landfill.

Parameter	Unit	Value	Average	SD	Standard effluent*
Temperature	°C	28-29	28.7	± 0.1	-
pH	-	5.32-6.21	5.82	± 0.2	6-9
COD	mg/L	895-910	902.5	± 110	150
Turbidity	mg/L	201-256	228.8	± 23	50
	µg/L	128-142	135	± 18	0.3

*Minister of Environment Regulation no. 5/2014 concerning waste water quality standards

2.1.2. *Coagulant.* Good quality of *Moringa oleifera* seeds (MOS) used were good provided by its characteristics as fresh (not rotten), matured, disease-free, brownish, and dried. The MOS was dried using an oven at 50 °C for 24 hours. 1 g of dried MOS was crushed using a food blender with 100 mL NaCl 1 M. The solution was then stirred for 30 minutes to then be filtered using a vacuum on membrane of 0.45 µm to obtain a solution of 1% MOS or 10,000 mg/L [24,27,29]. These concentrations have proven to be more effective in removal of turbidity, organic matter and heavy metals. Therefore, it is used for jar test trials in determining the optimum effective dose, pH, and optimum speed of rapid mixing.

2.1.3. *Constructed wetland.* The type of constructed wetland used in this study is vertical sub-surface flow constructed wetland (VSSFCW). VSSFCWs are made from glass of laboratory scale of 0.4 m x 0.4 m x 0.5 m. Then CWs in laboratory scale are filled with substrate. The substrate used must have good porosity to prevent clogging and have a special ability to absorb pollutants. Clogging is a fatal event in wetlands operation because it can reduce VSSFCW performance [1]. The substrate used is coarse aggregates, gravel medium, and zeolites. Aggregate coarse placement followed by gravel medium in VSSFCW bed aims to increase bio decomposition and mineralization [30]. At the bottom is placed zeolite to provide a wider area in the framework of the formation of biofilms so as to increase removal efficiency, especially organic matter [31-33]. Plants used in CWs in general have abundant availability locally and high efficiency, one of which is *Canna indica* [34]. Acclimatized plants use de-ionized water to provide initial nutrients for 21 days to be subsequently replaced by leachate landfills after 21 days. Changes are made by removing the water in the VSSFCW bed until it is completely dry. This was done so that the LLs to be added did not experience dilution when entering VSSFCW [1].

2.2. Methods

2.2.1. *Jar test trial.* Optimization of several factors is needed to obtain the best processing efficiency. Some determinants of flocculation-coagulation efficiency include type, dose, coagulant alkalinity, effluent pH, temperature, and mixing speed and time. In the optimization study carried out at a dose of coagulant and the speed and time of stirring so that the use of MOSP is cheap, able to remove colloids, and does not contain metal elements or reduce the alkalinity of the medium because it does not undergo hydrolysis makes this landfill leachate processing more effective [35].

2.2.2. *Determination of the optimum dose is done by preparing a jar test.* Preparation is done by placing the 500 mL beakers with distilled water for further drying. The landfill leachate is then removed from the refrigerator and left at room temperature. A total of 400 mL of landfill leachate was varied to 75, 100, 150, 175, 200, and 225 mg/L. Each dose was added to the beakers agitated by stirrer at 100 rpm for 4 minutes, then slowed to 30 rpm for 20 minutes and followed by sedimentation for 30 minutes. Raw landfill leachate and supernatant were then analyzed directly for COD content using the closed reflux method while Mn was analyzed using an atomic absorption spectrophotometer. Analyzes were also carried out for pH, temperature and turbidity. The optimum dose of MOSP is obtained at 175 mg/L.

2.2.3. Determination of the optimum pH is done by making a pH range of 5; 5.5; 6.0; 6.5; 7.0; 7.5; 8.0; 8.5; and 9.0 to find the optimum pH.

2.2.4. Optimization of flocculation-coagulation. The test is carried out by filling 6 beakers with a volume of 500 mL with a landfill leachate sample of 400 mL to be placed under the jar test apparatus. The speed of rapid mixing was varied to 100, 120, and 150 rpm for 6, 8, and 10 minutes while the speed of slow mixing was fixed at 30 rpm for 20 minutes with MOSP dose variations of 125, 150, and 175 mg/L. After 30 minutes' precipitation, the supernatant was filtered for Mn analysis as a control. The results obtained are compared to determine the optimum speed and time for rapid mixing. Optimum results are obtained at a speed of rapid mixing of 100 rpm for 10 minutes and slow mixing of 30 rpm for 20 minutes.

2.2.5. VSSFCW running. The optimum dosage, speed and time of the coagulation flocculation process with MOSP which provides the highest removal efficiency of various pollutants contained in landfill leachate are selected to flow towards VSSFCW. Landfill leachate which has undergone a process of flocculation-coagulation and deposition is analyzed its pollutant content before flowing to VSSFCW. Landfill leachate which has been separated from the dissolved solids is flowed intermittently with a discharge of 100 mL/sec. This means that the oxygen diffusion process when the bed surface is dry after the water has seeped into the lower layer is more optimal so that it can prevent clogging [36][37]. In addition, drainage of this type can increase removal of organics and nitrogen [38]. Observations were made with a variation of hydraulic retention time (HRT) for 5, 10, 15, and 21 days. The description of the series of experiments in this study is shown in Figure 1.

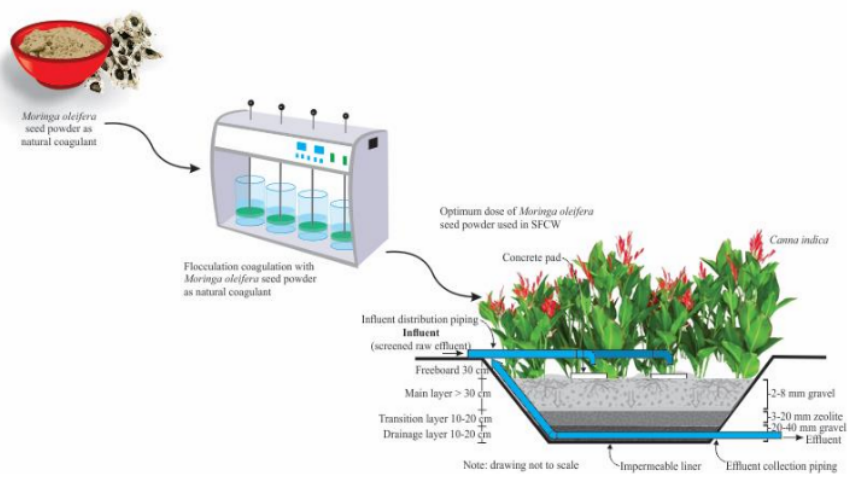


Figure 1. Schematic diagram of laboratory scale combined flocculation coagulation and constructed wetland.

2.3. Analytical and statistical analysis

Raw samples and effluents from each process (coagulation-flocculation and VSSFCW) were analyzed using standard methods for temperature, pH, COD and turbidity while heavy metals including Cd, Cr, and Fe used inductively coupled plasma mass spectrometers. All statistical tests were performed using Systat Version 8 by SPSS Inc. In all cases, significance was defined by $p < 0.05$. A two-way ANOVA was used to determine the significance of species and nutrient effects on biomass. Tukey's LSD was applied to the test for significance between treatment means.

3. Results and discussion

In the initial step, the optimum dosage of MOSP, pH, and speed and mixing time are determined. This was done to determine the composition of the MOSP dose, pH, and the speed and time of stirring which gave the removal efficiency of optimal COD, Turbidity, and Mn parameters. Determination of the optimum dose is done by varying the MOSP dose of 75, 100, 150, 175, 200, and 225 mg/L using a jar test. The most optimum MOSP to reduce the parameters of COD, turbidity, and Mn in LL is 175 mg/L. The addition of MOSP dose of 175 mg/L was able to achieve the percentage of removal for COD, turbidity and Mn respectively by 27.22%, 66.08%, and 92.01% as shown in Figure 2.

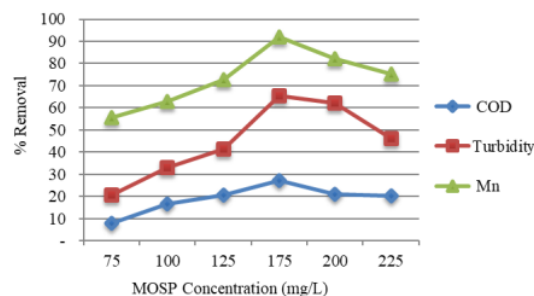


Figure 2. Effect of MOSP dose variation on COD removal, turbidity, and Mn.

Figure 2 shows that the addition of MOSP only gives slightly COD removal compared to turbidity and Mn. Despite the addition of MOSP concentrations, there was no significant increase in COD removal. In contrast to the effect of adding MOSP to COD, the turbidity obtained a fairly high removal efficiency but then a decrease occurs when the optimum dose is exceeded. This is because the excessive dose of coagulant will cause charge reversal and restabilizing of particles [39]. The highest removal efficiency is found in Mn where MOSP is able to act as a natural adsorbent for metals [40]. This is due to the activation of fat-soluble polyelectrolytes bound to metals to form complex formations [41].

The next variation is the optimum pH needed to achieve optimal results using MOSP as coagulant as shown in Figure 3. As seen in the optimum pH 6.5 a COD reduction of 32.16% was obtained while turbidity was 68.80% and Mn was 94.5%. The effectiveness of pH in the removal of various parameters of COD, turbidity and Mn due to the addition of MOSP as in Figure 3 occurs at pH 6.5. These results are in line with previous studies where the maximum removal due to the addition of MOSP in petroleum refinery processing occurs in the pH range 5-6.5 which results in removal efficiency below 82% [42] and in line with other previous studies [43]. This is due to the presence of basic amino acid proteins contained in MO acting as a proton acceptor so as to produce the release of hydroxyl functional groups and make the solution basic [42].

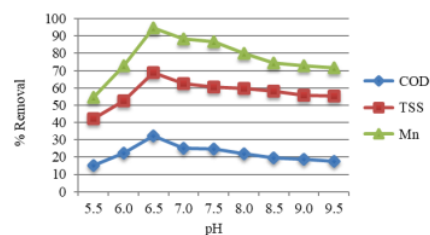


Figure 3. Effects of pH variations on removal of COD, turbidity and Mn.

In this study temperature variations were not carried out because they were expected to take place well at room temperature. In addition to variations in dosage and pH variations in the speed of stirring are also carried out as shown in Figure 4. This is done because the rapid mixing speed plays an important role in floc formation. This process serves to provide a place for rapid nucleation and deposition of MOSP. In this step, the MOSP coagulant will form a small flock to then form a denser flock with the size of the floc remained stable. After the sedimentation process, a denser flock will form a larger flock. Therefore we need a stirring speed that is conducive to the flocculation process in removing pollutants [44]. In this study the stirring speed obtained which gives the optimum percentage removal is 100 rpm for 10 minutes. Based on the results of the trial, the combined use of a MOSP dose of 175 mg/L was obtained at pH 6.5, the speed of rapid mixing of 100 rpm for 10 minutes and room temperature conditions.

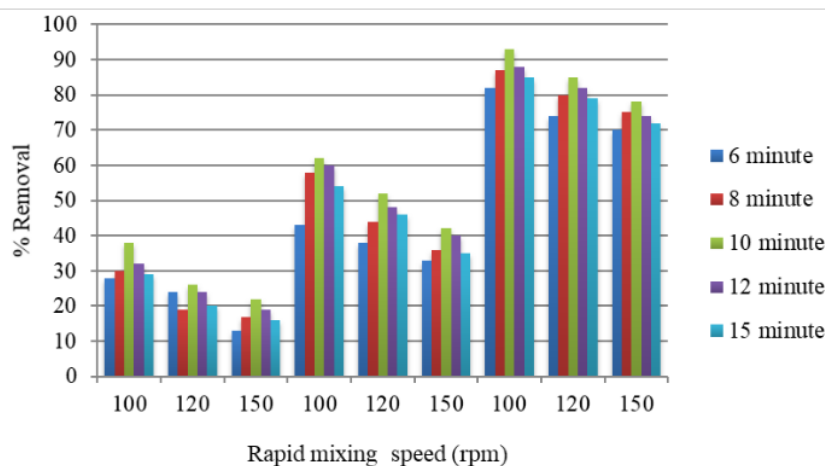


Figure 4. Effects of variations in stirring speed on the removal of COD, turbidity and Mn.

The results showed that at optimum conditions (pH 6.5, MOSP concentration of 175 mg/L, rapid mixing 100 rpm for 10 minutes and room temperature conditions) could improve the quality of LL by achieving the percentage removal of COD, turbidity and Mn respectively by 35.27%, 69.48% and 94.83%. Statistical tests were performed using the analysis of variance (Anova) to evaluate the significance of the difference between the average measured values. The results showed no significant difference for COD parameters but there were significant differences for turbidity and Mn ($P = 0.074$, $P = 0.0038$, and $P = 0.0026$ for COD, turbidity and Mn). As previously mentioned, this study only slightly percentage of COD removal is in line with previous studies which stated that MOS that did not undergo chemical modification process would result in a low percent removal of organic matter [45]. Other studies suggest that the use of MOS as a coagulant will cause high concentrations of COD [46]. This is because organic material does not experience precipitation in the coagulation process and is still present in the treated slaughterhouse wastewater. The organic material also causes an increase in BOD concentration [47]. Shan et al. also reported a dramatic increase in COD and BOD in treated wastewater due to the use of MOS as a coagulant, increasing oil content and leaving organic material on MOS cake [48].

The results showed that at optimum conditions (pH 6.5, MOSP concentration of 175 mg/L, rapid mixing 100 rpm for 10 minutes and room temperature conditions) could improve the quality of landfill leachate by achieving the percentage removal of COD, turbidity and Mn respectively by 35.27%, 69.48% and 94.83%. Statistical tests performed higher removal achieved for turbidity which was also in line with previous studies where the addition of MOS extracts of 10-30 mg/L at pH 6-8 was able to reduce turbidity by more than 90% [42]. Other research also states that the use of MO coagulant for

wastewater refinery processing can reduce turbidity by 12%-62.5% [26]. The decrease in turbidity by MO is caused by the process of absorption and neutralization of the positive colloid charge which binds the negative charge of the turbidity causing substances in wastewater [49]. The highest Mn removal obtained in this study was 94.85%. This is due to the addition of MOSP coagulant which then forms floc and attracts Mn metal into it. The addition of coagulants causes the transfer of material based on its nature, i.e., from what is not easily stored to be deposited either with or without an oxidation reduction reaction. The decrease in Mn content is probably due to the amphoteric MO protein binding to metal ions which have opposite charge which then causes the metal ions to precipitation [50].

The coagulation flocculation process using MOSP as a natural coagulant with the optimum dosage was able to reduce the COD content, turbidity, and Mn to 584.19 mg/L, 69.83 mg/L, 6.98 mg/L. This wastewater is processed by coagulation flocculation which will be treated in VSSFCW. The landfill leachate drainage process is carried out intermittently with a discharge of 100 mL/s. The drainage process is carried out intermittently with the aim of minimizing the occurrence of clogging in the CWs filter bed so as to allow the degradation of dissolved organic material that has accumulated on the media and allow the surface of the media to dry for a certain period of time. This type of drainage was chosen because it wanted to achieve a better aerobic process so that removal of organic material and ammonia nitrogen could be higher [51]. Observations were made with a variation of the hydraulic retention time (HRT) for 3, 5, 10, 15, and 21 days as shown in Figure 5.

In Figure 5, the optimum HRT is obtained for removal of the three parameters obtained on the 5th day. At that time, the percentage of removal achieved for COD, turbidity and Mn were 88.64%, 92.88% and 98.22%, respectively. That is because there was a decrease in the value of COD, turbidity and Mn from 584.19 to 66.36 mg/L for COD; 69.83 mg/L to 4.97 mg/L for turbidity and 6.98 mg/L to 0.12 mg/L for Mn. Furthermore, after day 5 there was a reduction in removal efficiency for all parameters. Hydraulic conditions can significantly influence the biogeochemical process, the composition of the biotic community, and the distribution of pollutants in constructed wetlands [52]. Hydraulic retention time is the time when there is contact between the plant and the substrate with the pollutant which is one of the most crucial things in determining the ability of efficiency and contaminant mechanism [53]. The long duration of HRT allows for higher removal of contaminants. This is due to the fact that wastewater does not move quickly to the outlet but has a longer contact with plant roots, media and microorganisms [37]. The results showed a significant difference between HRT variations and removal efficiency ($p < 0.005$).

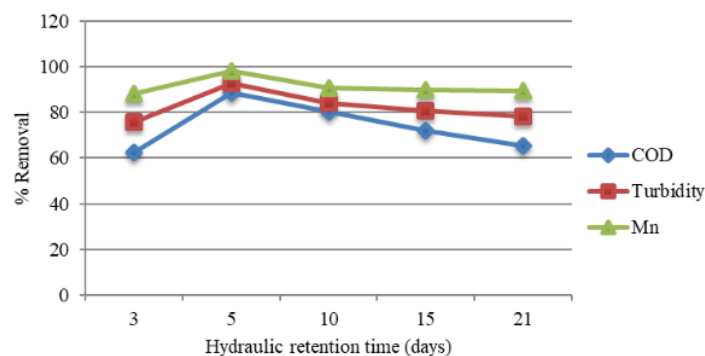


Figure 5. The effect of HRT variations on VSSFCWs on removal of COD, turbidity and Mn.

Figure 5 shows that VSSFCW are able to provide improved removal efficiency for all three parameters. This result is supported by an analysis of variance (Anova) which tests the significance of the difference

between the average measured values. The results show that there are significant differences for the parameters of COD, turbidity and Mn where a significant level of $p = 0.0028$ for COD, $p = 0.002$ for turbidity, and $p = 0.0012$ for Mn is obtained. In the removal process, the existence of *Canna indica* plays an important role in the effectiveness of processing with wetlands. Plants play an indirect role in creating conditions suitable for pollutant removal. For example, in the process of reducing COD, plants play a role as a place for attaching microorganisms that play a role in decomposing organic matter. The COD reduction process occur due to several processes namely the process of filtration, dissolved particle sedimentation, and anaerobic degradation of organic matter by microorganisms in constructed wetlands [54,55].

In contrast to COD removal, the turbidity removal process is caused by filtration and sedimentation by root macrophytes which reduce the distance between the media by forming the filter media to be tighter so as to reduce dissolved particles [56]. Likewise, in the removal of metals and nutrients where plants play a direct role. The process of Mn removal mechanism is through biological pathways, chemical precipitation and co-precipitation where binding of organic matter, sorption media and root surface of *Canna indica*, and suspended solids filtration by roots and media systems [57]. The contribution of each of these mechanisms depends on environmental conditions especially the pH and oxidation reduction potential. Therefore, the aboveground biomass must be harvested regularly [37].

4. Conclusion

This combination of technologies makes it possible to convert landfill leachate into an effluent that meets the ministerial environment regulation standard No. 5/2014 concerning waste water quality standards. The final result VSSFCW effluent is able to produce a COD value of 66.36 mg/L; 4.97 mg/L for turbidity and 12 mg/L for Mn where the respective standards set for COD, turbidity, and Mn are 150 mg/L, 50 mg/L, and 0.3 mg/L. These conditions can be achieved by using the coagulation flocculation process with MOSP under optimum conditions (pH 6.5, MOSP concentration of 175 mg/L, 100 rpm rapid mixing for 10 minutes) as pre-treatment. The composition was able to achieve the percentage removal of COD, turbidity and Mn respectively of 35.27%, 69.48%, and 93% with results that were still above the standard set by the government for COD and Mn of 584.19 mg/L for COD and 6.98 mg/L for Mn. Specifically, the turbidity of the coagulation flocculation processing has reached the government standard that is equal to 69.83 mg/L. Furthermore, the processing process was continued with VSSFCWs with the percentage of removal for COD, turbidity and Mn respectively at 88.64%, 92.88%, and 98.22%. The second stage of processing using VSSFCW is able to make the COD and Mn parameters meet the effluent standards set by the government.

17. Acknowledgment

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