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REMOVAL OF REFRACTORY COMPOUNDS FROM LANDFILL LEACHATE BY USING NANOFILTRATION

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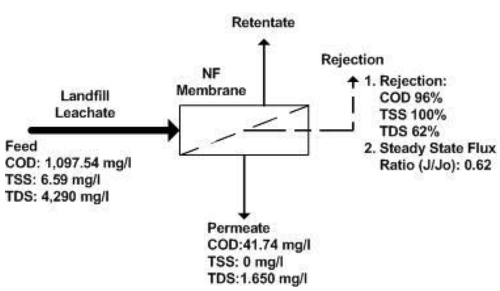
Graphical abstract



Abstract

Landfill leachate is a serious problem during treatment of municipal solid waste using landfill method. Less attention has been paid for the treatment of this leachate while this leachate is usually highly polluted. This study investigated the performances of nanofiltration membrane for treatment of landfill leachate (from Semarang, Indonesia). Landfill leachate was treated using NF99 nanofiltration membrane (pore size 200 Da). Synthetic leachate was used in this research which follows the characteristics of real leachate. Microfiltration (MF) membrane was used as a pretreatment before Nanofiltration (NF). The effect of pressure on membrane performance was observed. The membrane performance was examined for permeate flux and membrane rejection for TSS, TDS, and COD then compared to the effluent quality of existing leachate treatment. The rejection of COD, TSS and TDS were 96, 100 and 62%, respectively. The results suggest that the effluent had much better quality than the existing installation leachate treatment.

Keywords: Leachate, TSS, TDS, COD, Nanofiltration

1.0 INTRODUCTION

Due to increasing urban populations and the concomitant with changes in consumption habits, municipal solid waste has been identified as one of the most serious environmental problem which needs to be overcome immediately [1, 2]. In the solid waste management, landfill is one of the most prevalent methods used by many countries in the world due to its low cost [2-5]. However, landfilling in combination with percolating rainwater produces waste water known as leachate [6]. After the landfill is closed, leachate will continue to be produced for up to 30-50 years more [7]. Leachate from landfill is typically hazardous and heavily polluted wastewater [8-9]. Leachate disposal without treatment can cause serious environmental problems as it can percolate through soil and cause water contamination not only surface but also groundwater [6,8]. Therefore, to prevent environmental pollution caused by leachate, removal of harmful substances before entering the receiving waters is required [7].

Leachate has different chemical, physical and biological characteristics [6] and its composition depends on various parameters such as waste type, climatic conditions, type of operation, and age of landfill [10, 11]. Leachate may contain many organic compounds which are usually biodegradable. Nevertheles, it is also usually found refractory biodegrable consisting mostly ammonia-nitrogen, heavy metals, chlorinated organic and inorganic which usually contain soluble and suspended materials [1, 6-9, 12, 13]. Leachate also contains 0.2-1.5% solute in which the largest composition (80-95%) consists of a monovalent salt [14], and high COD concentrations [6].

Various leachate treatments have been proposed by using biological and physical-chemical processing. Often used biological treatment methods are sequencing batch reactor (SBR), trickling filter, rotating biological contactor (RBC), moving bed bio film reactor (MBBR), and anaerobic sludge blanket (UASB) [6]. Physical-chemical treatments are coagulation-flocculation method, chemical precipitation, ammonia stripping, and adsorption [1, 8, 11, 15]. However, these treatments require large areas, high operational and maintenance cost, long residence time, high energy use, and produce large amount of sludge [1, 15]. Therefore, an alternative technology for leachate processing is needed. One of the technologies that widely used in recent years is membrane technology [15].

Nanofiltration membranes are widely used in water and wastewater treatment [16-19]. This study aims to determine the performance of NF membrane in the removal of compounds COD, TSS, and TDS contained in the landfill waste water (leachate).

2.0 METHODOLOGY

2.1 Materials

Kaolin (Al_2O_3 $2SiO_4.2H_2O$) as TSS synthetic materials and NaCl for making TDS synthetic were purchased from local company CV Indrasari, Indonesia. Glucose ($C_6H_{12}O_6$) as material for producing COD synthetic was purchased from Merck, Darmstadt, Germany. Nanofiltration (NF99) membrane was donated by Alfa Laval, Denmark.

Real leachate obtained from Blondo landfill, Semarang was firstly characterized. The parameters examined were total suspended solid (TSS), total dissolved solid (TDS), and chemical oxygen demand (COD). The results of leachate characterization were TSS 178 mg/L, TDS 4,305 mg/L, and COD 1,781.4 ppm. Then synthetic leachate wastewater was prepared based those characterization results.

Synthetic TSS was made by dissolving 178 mg of kaolin into 1L of distilled water. Synthetic TDS (4,305 mg/L) was produced by dissolving 4.384 grams of NaCl to 1 L distilled water. To produce 1,781.4 ppm of synthetic COD, the first thing to do was by calculating COD concentration in a 1,000 mg/L glucose solution. The resulted test in laboratory showed that the value of COD for 1,000 mg/L of glucose was 1,212 mg/L. The glucose needed to make 1781.4 mg/L COD was 1,469.8 mg. This value was calculated by using equation bellow:

$$\frac{1{,}000\frac{mg}{L}glucose}{1{,}781.4\frac{mg}{L}glucose} = \frac{1{,}212\frac{mg}{L}COD}{x}$$

2.2 Method

Before used, all the membranes with effective area of 13.8 cm^2 were soaked in distilled water and then compacted. Thereafter crossflow filtration was performed. The pressure was varied (4, 5, and 6 bars). The series of filtration tools used are shown in Figure 1.

The permeate flux of membrane, J (L/m^2 h) at different pressures was determined using equation (1).

$$J = \frac{V}{A\,x\,t} \qquad (1)$$

where V is the volume of permeate (liters), A is the surface area of the membrane (m^2), and t is the time required (hour). Percent rejection (R) was calculated using equation (2), where Cp is permeate concentration and Cf is feed concentration.

$$R = 1 - \frac{C_p}{C_f}x100\% \qquad (2)$$

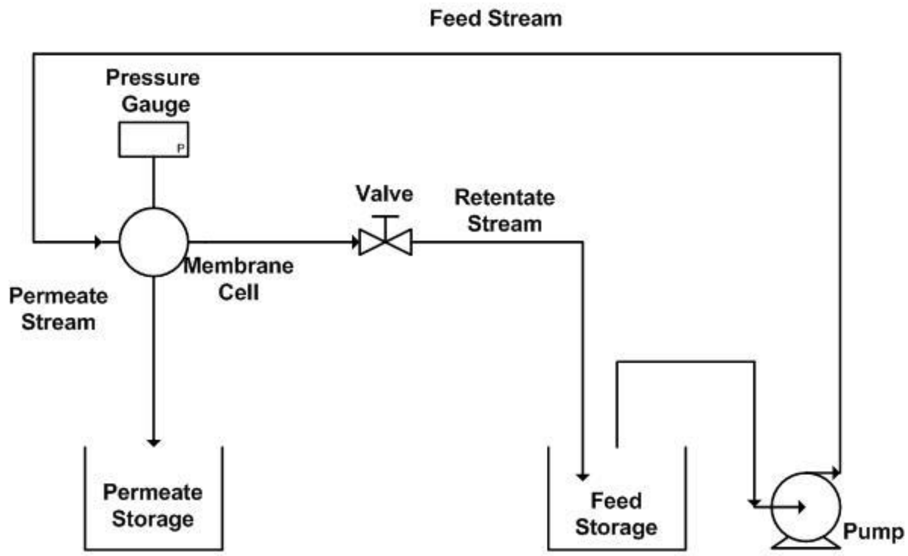


Figure 1 Design of membrane filtration

Permeate concentration for TSS parameter was tested by using turbidimeter, TDS using TDS meter, and COD using spectrophotometer. The membrane morphology was visualized using a Scanning Electron Microscope/SEM (JEOL JSM-6510LA SEM, Japan).

3.0 RESULTS AND DISCUSSION

3.1 Leachate Characterization

Raw leachate taken from Blondo Landfill, Semarang, Indonesia was firstly characterized. The result is presented in Table 1.

Table 1 Characteristics of landfill leachate

Parameter	Unit	Characteristics of leachate	Quality standards (QS)*	Informa-tion
Physical Parameters				
Temperature	°C	27	38	
TSS	mg/L	178	100	>QS
TDS	mg/L	4.305	2.000	>QS
Chemical Parameters				
pH		8,40	6,0 – 9,0	<QS
Iron (Fe)	mg/L	9,80	5	>QS
Copper (Cu)	mg/L	0,11	2	<QS
COD	mg/L	1.781,40	100	>QS

*Source: *Regional Regulation of Central Java Province Number 5 the year 2012 concerning the Quality Standard of Wastewater for Uncertain Business and/or Activity of its Standard of Quality*

It can be clearly seen that all parameters, i.e., TSS, TDS, and COD are beyond the regulatory limit set by Government of Indonesia. Therefore, this study is focused on the elimination of these three parameters.

Due to the high concentration of organic compounds in the leachate [7], it allows the formation of fouling in the membrane filtration process [20]. Leachate from landfill has diverse characteristics that will affect membrane performance. Therefore, the use of synthetic leachate is necessary to determine the membrane performance in the removal for each or mixed parameters.

3.2 Pre-Treatment Leachate Using Microfiltration Membranes

According to Mojiri [2] on leachate treatment, microfiltration (MF) can be used as an initial treatment that can be combined with other membrane processes. In addition, Yao [21] has also explained that MF is an effective method for removing colloids and suspended solids. The rejection rate of the MF membrane for dissolved solutes is still not significant, so it is often used as pre-treatment for advanced processing using other membrane processes such as ultrafiltration (UF), nanofiltration (NF), or reverse osmosis (RO).

In the leachate treatment using MF membrane, the parameters are excluded not only TSS but also other parameters such as TDS, and COD. Thus, it is necessary to analyze the resulting flux differences and rejection rates for each single solution (TSS, TDS, COD) and mixed solutions. The graph of flux value and rejection rate can be seen in Figure 2 and Table 2.

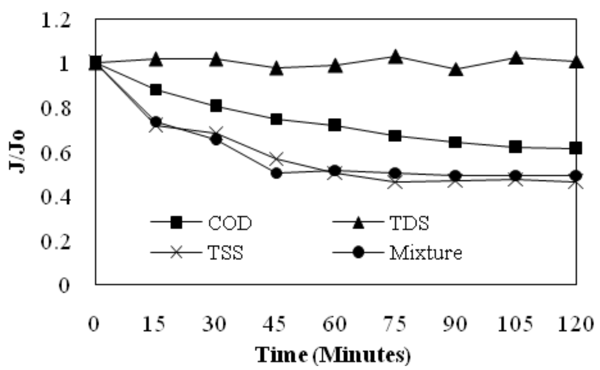


Figure 2 Normalized flux profile with time using MF with Jo = 590.14 L/m².h (P = 1 bar)

Figure 2 shows the flux profiles of parameters which are removed from leachate using MF membranes. The flux of the COD solution, TSS solution, and mixed solution of the four parameters decreased at the 15th minute to 75th minutes, and then the flux was stable. It is also seen that there is no significant decrease of normalized flux in TDS removal. While on the other parameters, the flux decreased at a certain minute. Due to absence of fouling formed on the surface of the membrane there was no significant flux reduction in TDS removal. In general, three stages of flux reduction occurred in the MF membrane, (a sharp decrease, gradual decline, and ended by steady-state flux). The statement is in accordance with the results obtained in previous study [22]. The decrease of flux showed that there was fouling on the MF membrane caused by accumulation of particles on the membrane surface.

Table 2 Concentrations of parameters on microfiltration membranes

Parameter	MF					
	Single Synthetic leachate			Mixed synthetic leachate solution		
	Concentration (mg/L)		Rejection Rate (%)	Concentration (mg/L)		Rejection Rate (%)
	Feed	Permeate		Feed	Permeate	
COD	1.768	1.539	13	1.899	1.163	39
TDS	4.330	4.250	2	4.410	4.290	3
TSS	178	6,58	96	178	6,59	96

Table 2 showed the difference of rejection rate for each parameter. The highest rejection rate indicated by TSS removal which reached 96%. While for the removal of TDS, and COD were 3%, and 39%, respectively. The MF membrane has microporous structure, and the particle separation process takes place based on pore size. Therefore, many separation processes occur on the outer surface of the membrane [23]. TSS solutions are made from kaolin which has a particle diameter size of 0.1 to 2 µm [24] while the pore size of the microfiltration membranes used is 0.8 µm. Thus, most kaolin was retained on the membrane, caused high rejection of TSS and caused significant reduction flux due to fouling.

Meanwhile, the molecular weight of COD (glucose), and TDS (NaCl), are 180.2 g/mol; and 58.5 g/mol respectively [17,25]. So, these parameters (COD and TDS) have low rejection rates on MF, because the particle size and molecular weight for these parameters are smaller than the pore size of the MF membrane. Consequently, most these pollutants pass through membrane pores resulting low rejection rate.

3.3 Effect of Operating Pressure on Nanofiltration Membrane Performance

The effect of pressure on membrane performance was studied. Membrane performance can be determined from the flux profiles shown in Figure 3, Figure 4 and Figure 5. Meanwhile, rejection rates were shown in Table 3 and Table 4.

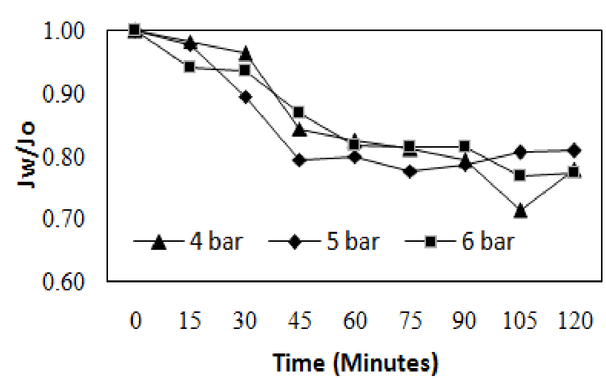


Figure 3 Time-normalized flux profiles in TSS solution (Jo 4 bar = 45,19 L/m².h, Jo 5 bar = 51,98 L/m².h, Jo 6 bar = 87,13 L/m².h)

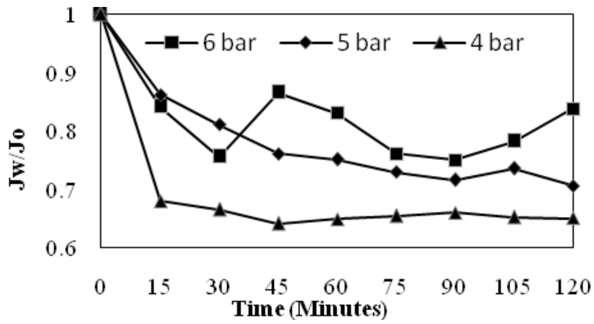


Figure 4 Time-normalized flux profiles in TDS solution (Jo 4 bar = 52,83 L/m2.h, Jo 5 bar = 48,15 L/m2.h, Jo 6 bar = 83,79 L/m2.h)

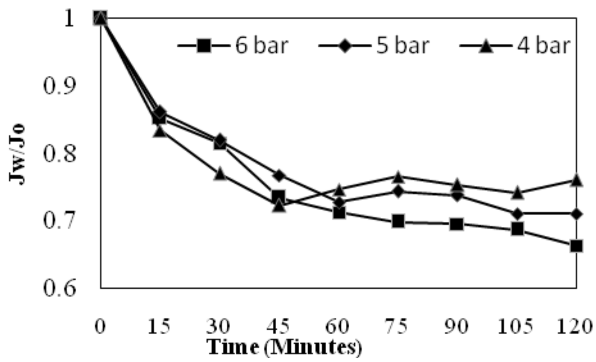


Figure 5 Time-normalized flux profiles in COD solution (Jo 4 bar = 40,32 L/m2.h, Jo 5 bar = 55,13 L/m2.h, Jo 6 bar = 68,03 L/m2.h)

Figure 3-6 shows that the decrease in flux profile significantly occurred in the 15th minute of experiment then it remained stable at the 75th minute for TSS, and COD solution. Whereas for TDS and mixed solutions, the flux was stable in the 45th minute. This reduction of flux profile of nanofiltration membrane could be due to fouling [26] and concentration polarization [27]. According to Schafer *et al.* fouling is an irreversible process that results in decreased membrane performance due to deposition of suspended substances or solutes on membrane surface [20]. While the concentration polarization occurs due to the accumulation of solute that is retained on the membrane surface and reversible [27].

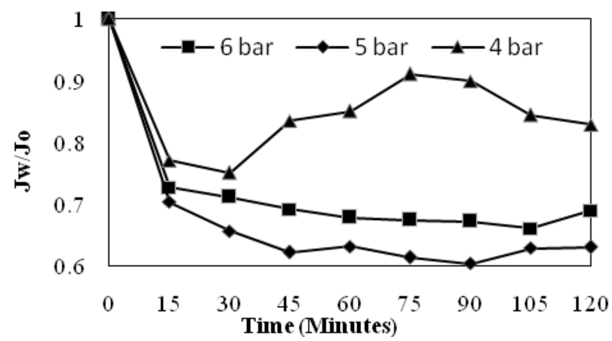


Figure 6 Time-normalized flux profiles in mixed solution(Jo 4 bar = 42,80 L/m2.h, Jo 5 bar = 68,89 L/m2.h, Jo 6 bar = 72,33 L/m2.h)

Table 3 Concentration of permeate and rejection rate of single synthetic leachate solution at various operating pressures

Parameter	Feed Concentration (mg/L)	Single Synthetic Solution					
		4 bar		5 bar		6 bar	
		C (mg/L)	R (%)	C (mg/L)	R (%)	C (mg/L)	R (%)
TSS	6.58	0	100	0	100	0	100
TDS	4,250	2,880	32	2,480	42	2,490	41
COD	1,392.29	78.63	94	71.94	95	69.36	95

Table 4 Concentration of permeate and rejection rate of mixed synthetic leachate solution at various operating pressures

Parameter	Feed Concentration (mg/L)	Mixed Synthetic Solution					
		4 bar		5 bar		6 bar	
		C (mg/L)	R (%)	C (mg/L)	R (%)	C (mg/L)	R (%)
TSS	6.59	0	100	0	100	0	100
TDS	4,290	1,680	61	1,650	62	1,840	57
COD	1,097.54	34.69	97	41.74	96	42.46	96

The removal of TSS, TDS, and COD as a single or mixed solution are shown in Table 3 and 4. Rejection is resulted by separation process that take place on the membrane. Table 3 shows that the rejection rate of COD for all variation of pressures for a single synthetic solution was ranging from 94-95%. While Table 4 shows the rejection rate of synthetic mixture solution that reached ± 96%. This result was in accordance with Madaeni, and Mansourpanah [28]. They reported that COD removal could reach 98% by using nanofiltration membrane.

The permeate concentration in Table 3 was then compared with Regional Regulation of Central Java Province, number 5 year 2012 concerning the quality standard of wastewater. All parameters tested on three operating pressures, both single and mixed feeds, and permeate quality was produced which met the quality standard for TSS, and COD parameters.

The results of this study indicated that the operating pressure of the nanofiltration membrane does not have a significant impact on the rejection of the three types of pollutant parameters. This is slightly different from the results reported by Mohammad *et al*. [16]. His result showed that the greater the operating pressure the higher efficiency of pollutant removal nanofiltration membranes. Overall, these results indicate that in the pressure 4 bar, NF99 showed its best performances in decreasing TSS, TDS, and COD.

According to Galanakis *et al*. [17], charge exclusion and sieving effects are two main factors that influenced separation process on nanofiltration membrane. Charge exclusion depends on three parameters: (1) membrane charge that depends on the pH of the feed solution, (2) ionic strength, and (3) ion valence [17]. While sieving effect took place due to the difference in pore size between membrane pores and dissolved particles in the feed solution [23]. The separation process of neutral feed solutions is caused by sieving mechanism [16].

The separation of salts by nanofiltration membranes is more complicated and can occur in combination based on molecular size and the Donnan exclusion effect (electrostatic interaction) [29]. The electrostatic interaction between solution and membrane is influenced by pH value of feed solution and the isoelectric point of membrane. The pH of the feed solution was 5 whereas the isoelectric point of the NF99 membrane is close to 4 [17] resulted negative charge of the NF99 membrane. The negative charge of NF99 caused by pH of TSS solution which is above the isoelectric point of the NF99 membrane. Synthetic TSS solution was made from negatively charged kaolin so when the TSS solution is contacted with a negatively charged NF99 membrane, there will be a refusion. Therefore, it is difficult for particles to pass through membrane and it resulted in high percentage TSS rejection. After filtration process using nanofiltration membrane, the concentration of TSS solution is lower, and rejection rate reached 100% for all pressure variations. The removal of TSS occurred due to the interaction between ion and membrane surfaces.

The separation mechanism of the TDS solution is similar to the separation in the TSS solution. A synthetic TDS solution pH 5 was prepared from technical NaCl comprising Na+ and Cl- ions. Because pH of TDS solution was 5, so the surface of NF99 have negative charge. According to Tu [29], salt separation process in the membrane occurred because membrane rejects the co-ion (ion having the same charge as the membrane surface) for solution electroneutrality, the counters (ions whose charge unequal with the membrane surface charge) in this case are Na+ also rejected by the membrane, so salt retention took place. Rejection rates in single or mixed synthetic TDS solutions decreased at 6 bar because the driving force was greater, allowed foulant to pass through the membrane and resulted lower TDS concentration in permeate.

The synthetic COD solution used in this study was made from uncharged glucose material. Thus, the elimination process may occur due to sieving mechanism. In this separation, the particle size which larger than the pore size of the membrane will be retained on the membrane surface while the smaller particle size will pass through membrane.

3.4 SEM Analysis

SEM was used to determine membrane morphology at the top layer of NF99 nanofiltration membrane. Tests were performed on fresh membranes, and membranes after treatment using a mixed synthetic leachate solution with 2,000x magnification can be seen in Figure 7.

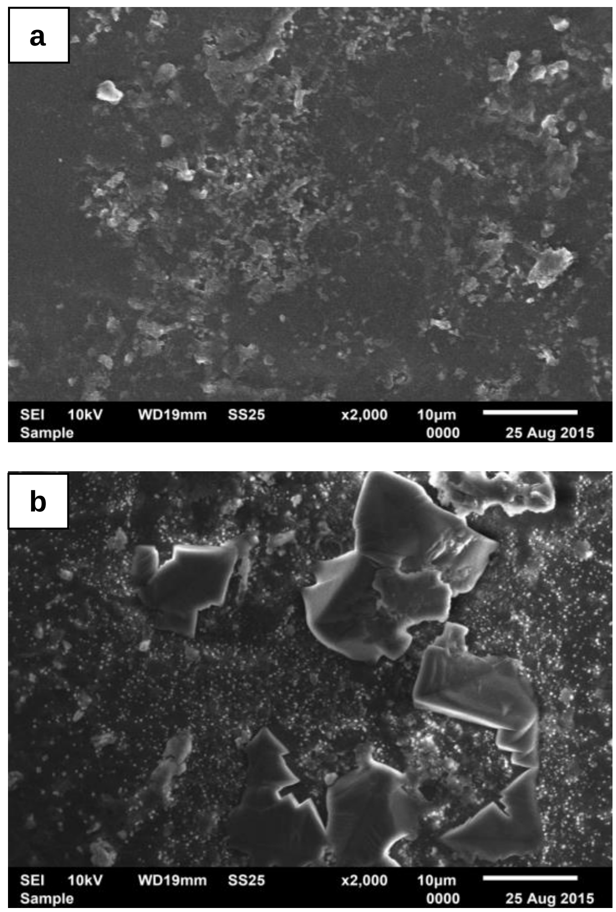


Figure 7 SEM results on the surface (a) fresh membrane, membrane after treatment using (b) mixed synthetic leachate solution

In Figure 7 the fresh membrane NF99 (a) has relatively smooth membrane surface morphology and no fouling was observed. This is because the membrane is still clean, has not been used for filtration, so there is no impurity on the surface of the membrane. After filtration process using synthetic leachate solution (b) the cake layer on the membrane surface was clearly seen.

There are many factors causing cake formation on the membrane surface. One of them is the suspended solids that still have not been removed from membrane. Based on the size of colloidal particles and membrane pores, colloidal fouling might occur due to the accumulation of particles on the membrane surface forming cake layer or it penetrates the membrane pores [30]. For solutions containing organic compounds where only few suspended solids available, can cause adsorption of organic compounds on the membrane surface [31]. The presence of other organic and inorganic compounds contained in landfill leachate also plays an important role in the formation of fouling because it allows interaction between existing components. The possible interactions are organic compounds with colloids, organic compounds with metals, and metals with colloids.

4.0 CONCLUSION

The removal of TSS, TDS, and COD in both single and mixture synthetic leachate has been studied with various pressures using MF membranes as pretreatment and NF99 membranes as main treatment. The optimum operating pressure to remove TSS, TDS, and COD contained in leachate was 4 bar. Synthetic leachate derived from the Blondo landfill can be remove using nanofiltration membrane. The TSS removal reached 100%, TDS 62%, and COD 97%.

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