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REMOVAL OF SOME CONTAMINANTS FROM PRE COAGULATED SEMI-AEROBIC LEACHATE USING MEMBRANE FILTER

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Abstract: Membrane filtration process for water treatment has been well established. However, its application on leachate effluent treatment after coagulation process is quite limited. The present paper investigated the suitability of the coagulation process on leachate effluent treatment generated from the Pulau Burung sanitary landfill site. Three types of membrane were used in this study, namely, nylon, resin and poly-propylene (PP). The pore sizes of membranes were 1µm and 5 µm. The effects of different filtration rates on leachate treatment were studied. The parameters studied were chemical oxygen demand (COD), colour, suspended solids (SS), and ammoniacal nitrogen (NH_4N). Poly-aluminium chloride was used as a coagulant in the coagulation and flocculation processes. Results indicate that PP membrane with pore size 1µm gave the best performance in reducing all the parameters, whereas the nylon membrane gave the poorest results. The filtration rate of 100mL/min exhibited the highest removal efficiency. PP membrane with pore size of 1µm at a filtration rate of 100mL/min is the most effective membrane to remove COD, colour, SS and NH4N.

Keywords: Membrane filtration, Leachate, Poly-propylene, Nylon, Resin

1. Introduction

Landfilling is the most common method for municipal solid waste disposal adopted by different communities in Malaysia (Agamuthu, 2001; Omran et al., 2007; Omran et al., 2009). The reason is landfilling is considered the simplest, and cheapest among the disposal methods in many areas (Agamuthu, 2001; Omran and Gavrilescu, 2008; Omran et al., 2018). Landfilling is also advantageous in that the holes produced from quarries and mineral workings are filled with waste materials to produce restored landscapes (Williams, 2005). However, one of the typical problems of landfilling methods is leachate generation. Leachate migrates from landfills, causing serious pollution in the groundwater aquifer and adjacent surface water. Consequently, one of the major issues that should be dealt with is the collection, storage, and implementation of a suitable treatment for highly contaminated leachate. Therefore, biological treatment and physicochemical treatment such as activated sludge treatment, rotary disc, sedimentation with coagulation, activated carbon adsorption, and membrane processes, have been widely used in leachate treatment. These methods can remove or reduce chemical oxygen demand (COD), biological oxygen demand (BOD), metals, suspended solids, colour, and nitrogen (Agamuthu, 2001; Aziz et al., 2010a). Coagulation and flocculation process are widely used in water, and wastewater treatments and these techniques are important for the treatment processes (Amokrane et al., 1997, Prakash et al., 2014; Aziz et al., 2010b). The coagulation process is effective for removing high-concentration organic pollutants, heavy metals, and some anions (Kreith, 1994). The characteristics of leachate to be treated before and after coagulation are shown in Table (1).

Table 1. Characteristics of leachate before and after coagulation and membrane filtration

Raw leachate		After coagulation		After 4 days membrane filtration using 5µm PP at 100 mL/min		After 4 days membrane filtration using 1µm PP at 100 mL/min	
Parameter	Concentration	Removal (%)	Concentration (mg/L)	Removal (%)	Concentration (mg/L)	Removal (%)	Concentration (mg/L)
COD (mg/L)	528	40	317	59.4	128	64.8	111
Colour (PtCo)	1970	75	493	85.2	73.1	87.6	61.2
SS (mg/L)	406	77	93	99.5	0.5	99.5	0.5
NH_3-N (mg/L)	245	22	191	51.7	92.3	52.7	90.3

Source: Hwang et al. (2009)

Currently, poly-aluminium chloride is widely used in electroplating waste treatment because of its relatively low molecular weight and a high cationic charged. Polymeric coagulants also have some ability to cause flocculation. Flocculation by polymer coagulants is caused by their molecular weight, which is very high compared with that of inorganic coagulants (introduction to Environmental Engineering Website). Membrane is a type of separation process utilizing a permeable thin pliable layer that acts as a boundary, lining, or a partition to separate impurities from streams (Md Isa et al., 2004). In the membrane process, the application of shear-enhanced filtration is a technique that separates the impurities (i.e., suspended or dissolved materials, or waste materials) according to molecular weight and size. The application of pressure within the system causes the membrane to serve as a sieve; thus, membrane construction engulfs thousands of pores within the surface area. The current research concentrates on the leachate generated from the Pulau Burung Landfill Site (PBLS) in Penang, Malaysia. PBLS is located at the southern district of Seberang Perai. It was constructed by Municipal Council of Seberang Perai (MCSP) in 1980. PBLS encompasses a total area acre, where both municipal and industrial solid wastes from Seberang Perai were deposited indiscriminately. This site receives 1500 tons of solid waste daily (Aziz et al., 2004). Leachate is collected in the retention ponds and is recycled back to the landfill site to increase the decomposition of wastes. In the rainy season, the quantity of leachate in retention ponds and landfill leachate treatment plant increases dramatically, hence, leachate flowing out into the drainage system of the area is possible. The MCSP and other surrounding residences are concerned about this matter. As PBLS is a semi-aerobic landfill, it produces less concentrated pollutants; therefore, the use of membrane technology to treat the leachate effluent is possible. The objective of this study is to apply the poly-aluminium chloride (PAC) as a coagulant to the sample using the recommended experimental conditions of Ghafari et al. (2005; 2009). The goal of this process in leachate treatment is to study the removal efficiency of COD, SS, colour, and NH_4N in leachate effluent by membrane filtration using three types of membrane filters: (1) poly-propylene (PP) at 1µm and 5µm, (2) nylon and (3) resin and at different filtration rates.

2. Materials and Method

2.1 Leachate collection and previous procedures

Leachate samples were collected from PBLS from November 2008 to February 2009 (Table 2). If the tests could not be conducted on the same day, the samples would be stored in a refrigerator and maintained at 4°C. Coagulation and flocculation studies were performed in a standard jar-test apparatus (Jar Tester Model CZ150) consisting of six paddle rotors (24.5mm x 63.5 mm) equipped with six beakers at 1 L volume each. Leachate samples were removed from the refrigerator and conditioned for about 3 h under ambient temperature. Samples were agitated thoroughly for the re-suspension of settled solids prior to the tests conducted (Aziz et al., 2007; 2004). The chemical reagents used as coagulants was poly-aluminium chloride (PAC) conducted at 1.9 g/L PAC in the highest removal at pH 7.5 under 80 rpm rapid mixing for 45 s followed by 15 min slow mixing at a speed of 40 rpm at a settling time of 30 min. These factors are based on recommended experimental conditions of Ghafari et al. (2005; 2009).

Table 2. Characteristics of Leachate from Pulau Burung Landfill Site (PBLS)

Parameter	25/11/08	22/12/08	14/1/09	17/2/09
COD (mg/L)	508	529	540	534
Colour (PtCo)	1960	1820	1700	2400
SS (mg/L)	216	343	600	465
NH	269	246	238	226

2.2 Membrane filtration reactor

The effluent was then transferred to the membrane filtration reactor with a tank size of 350 mm × 550 m × 400 mm. As the system used gravity flow, the bottom of the tank was drilled, and the hole was connected to a pipe. To increase the pressure head of the leachate effluent, the reactor was placed on a table (Figure 1). The pipe was connected to a valve used to control the filtration rate of the leachate effluent. At some turning points, the connection was made by using the standard elbow of 90°. The flow system was then connected to a membrane filter used to treat or filter the sample. After the filtration process, a container was used to collect the effluent. At the same time, a pump was used to transfer the effluent from the container to the tank to complete the cycle (Figure 2).

Figure 1. Front and Side Views of the Membrane Filtration Reactor

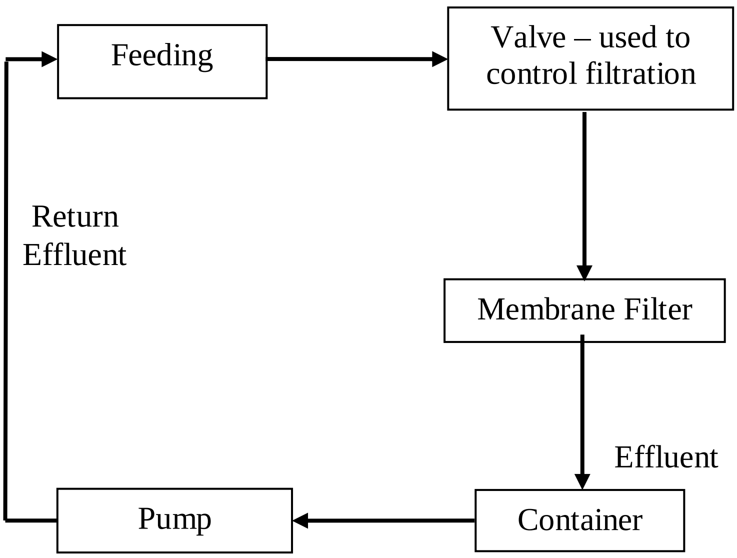

Figure 2. Diagram of the Experimental Set up of the Reactor

Phase I: Different Filtration rate for PP

The first part of the research discussed the effect of filtration rate on removing suspended solids (SS), colour, ammoniacal nitrogen (NH_4N), and chemical oxygen demand (COD). In this aspect, the research only covered the filtration rate range from 100-140 mL/min. A poly-propylene membrane with a pore size of 5 m was used in the current study. Each filtration rate ran continuously for 4 days in a week. Daily changes in each parameter were determined. The removal percentage of each parameter for different filtration rates was also determined at the end of each week.

Phase II: Different types of membrane filters

The second part of the research was to investigate the efficiency of the different types of membrane filters. The three types of membranes used in the current study were nylon, resin, and poly-propylene (PP) with a pore size of 1 and 5 µm. The effect of pore size on each membrane was also studied by comparing the results obtained from the PP (1 µm and 5 µm) membrane. A constant flow rate of 100 mL/min was used in this study. Each membrane filter ran continuously for 4 days in a week. Daily changes in each parameter were determined at the end of each week.

Phase III: Data Analysis

The removal percentage of each parameter was determined in each type of membrane filter. The COD was measured based on the "5220 D. closed reflux colorimetric method" from the standard methods for the examination of water and wastewater (APHA, 1992). SS detection was conducted using the DR/2010 spectrophotometer through the photometric method (non-filterable residue), method 8006 (Hach Company Procedure, 1998). The SS was measured at 810 nm wavelength (program number 630). The unit of SS is in mg/L, in the range of 0-750 mg/L. NH_4N was determined at 425 nm using a DR/2010 spectrophotometer through the Nessler method, method 8038 (Hach Company Procedure) adapted from the standard methods for the water and wastewater tests. The light yellowish colour produced by the Nessler-ammonia reaction was then measured at a 425 nm wavelength of and presented in mg NH_4N/L. Colour was measured by the APHA platinum-cobalt (PtCo) standard method, method 8025 (Hach Company Procedure, 1998), which was adapted from the standards methods for water and wastewater tests using DR/2010 spectrophotometer. Colour was measured at a 455 nm wavelength (program number 120) in the range of 0-500 PtCo. Prior to each determination, the samples were filtered using filter paper with a pore size 0.45 µm; distilled water was used as a blank. Statistical analysis was conducted using ANOVA and the MINITAB 14 Release software. A one-way ANOVA tested the hypothesis that the means of several populations or two groups are equal or the same by extending the two-sample t-test, in where the population variances are assumed to be equal. Factor levels were selected from different values. The analysis of each factor level should correspond to a larger population with its own mean. The estimation of the mean level is the sample mean for the whole population. The design of the one-way ANOVA is based on (Eq.1):

$$Y = \mu\varepsilon \qquad (1)$$

Where Y = observation vector, µ = vector of the means composed of the treatment means, and ε = error vector.

3. Results and Discussion

3.1 COD Removal

The COD was 40% prior to the filtration process. The removal of COD at a filtration rate of 100 mL/min was 59.4%. COD removal at 110, 120, and 130 mL/min filtration rates was higher than that at 60%, which was 65.7%, 67.3%, and 64.6%, respectively. However, the percentage of COD removal dropped to 55% at a filtration rate of 140 mL/min (Figure 3). Leachates from municipal solid waste landfill sites are usually defined as hazardous and heavily polluted wastewaters (Barbusinski & Pieczykolan, 2010). Leachates contain a large amount of organic matter (i.e., biodegradable, but also refractory to biodegradation), in which the humic type of constituents in an important group (Kang et al., 2002), as well as ammonia nitrogen, heavy metals, chlorinated organic and inorganic salts (Wang et al., 2002). According to Vogel et al. (2000), one- third of the COD in leachate is influenced by the inorganic matters such as Fe (II), manganese (II), sulphide, ammonia and chloride. As these types of materials can react with PAC to form sludge; they can easily filtrate by micro-membrane. Therefore, COD removal is high in the case of the simple membrane filtration process.

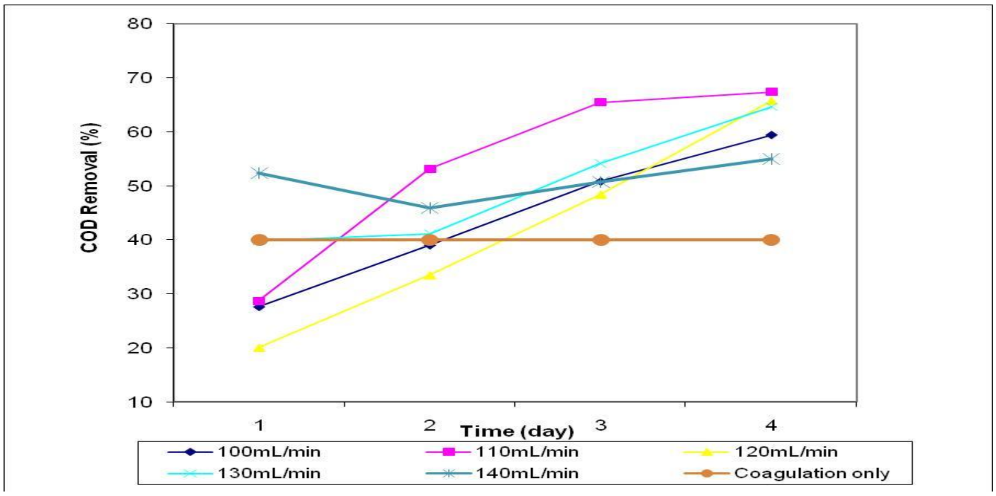

Figure 3. COD Removal versus Time at Different Filtration Rates using 5µm PP at pH 7.5

3.2 Colour removal

Figure (4) shows the results of colour removal. Before the filtration process, colour removal was 75%. Colour removal at all filtration rates was high in the range of 82%-90%, higher than the removal for COD. Colour removal at 100 mL/min filtration rate was 89.4%, and 82% at the filtration rate of 130 mL/min. Colour is used as an identifier of secondary pollutants in wastewater, although no record shows that it can affect human health (Coro & Laha, 2001). However, the presence of colour in water causes a negative water perception. According to the Environmental Protection Agency in 1996, the minimum standard of colour in drinking water is 15 colour units. The presence of organic matters such as humic substances in water also contributes to the colour of water. Zouboulis et al. (2004) pointed out that the decomposition of organic matter such as humic acid, causes water to become yellow, brown or black. Colour is caused by turbidity is known as true colour, whereas colour caused by the presence of ion metallic or suspended solids in water system is known as apparent colour (Standard Method for Water and Wastewater, 1984). As such, the removal of suspended solids and turbidity also influences the removal of colour. The SS in the current study was high; therefore, the overall result for colour removal was also very high at 80% and above. This result may be due to the presence of ion metallic or organic dissolved matters, such as humic substances (Zouboulis et al., 2004) in leachate effluent, which can pass through the membrane filter that has undergone coagulation and flocculation processes to form sludge. This sludge was formed after filtering by the membrane filter. Thus, the current study obtained a better result for colour removal.

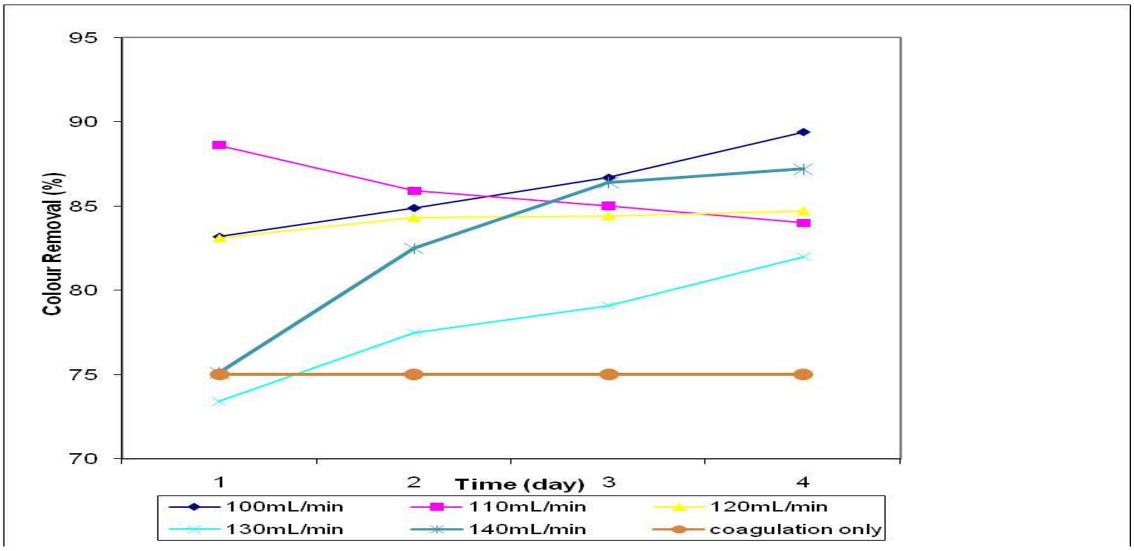

Figure 4. Colour Removal versus Time at Different Filtration Rates by Using 5µm PP and pH7.5

3.3 Suspended solids removal

Figure (5) shows the SS removal at different filtration rates. The removal of SS was 77% prior to the filtration process. SS removal at the filtration rate of 130 and 140 mL/min was slightly lower (about 97.7% and 98.9%, respectively) than that of others. At the filtration rates of 110 and 120 mL/min, the removal was almost the same at the end of the experiment. High SS removal was observed at the filtration rate of 100 mL/min, with 99.5% reduction. In conclusion, a low filtration rate provides high results in SS removal because of higher retention time.

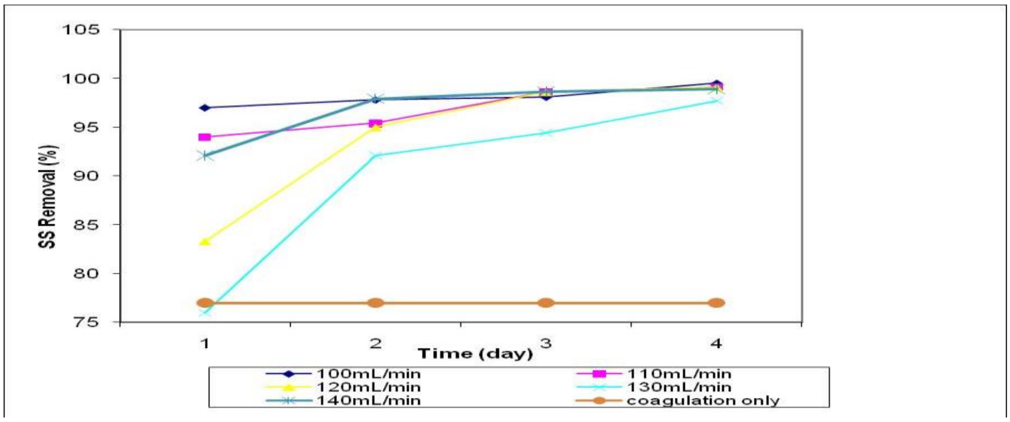

Figure 5. Suspended Solids (SS) Removal versus Time at Different Filtration Rates by using 5µm PP and pH7.5

3.4 Ammoniacal Nitrogen (NH_4N) removal

The removal of NH_4N was 22% prior to the filtration process. The percentage of NH_4N removal at all filtration rates was low and was in the range of 40%-55%. Organic matter in leachate highly contributes to NH_4N. Therefore, NH_4N of the leachate also decreased once the organic matter was removed (Figure 6).

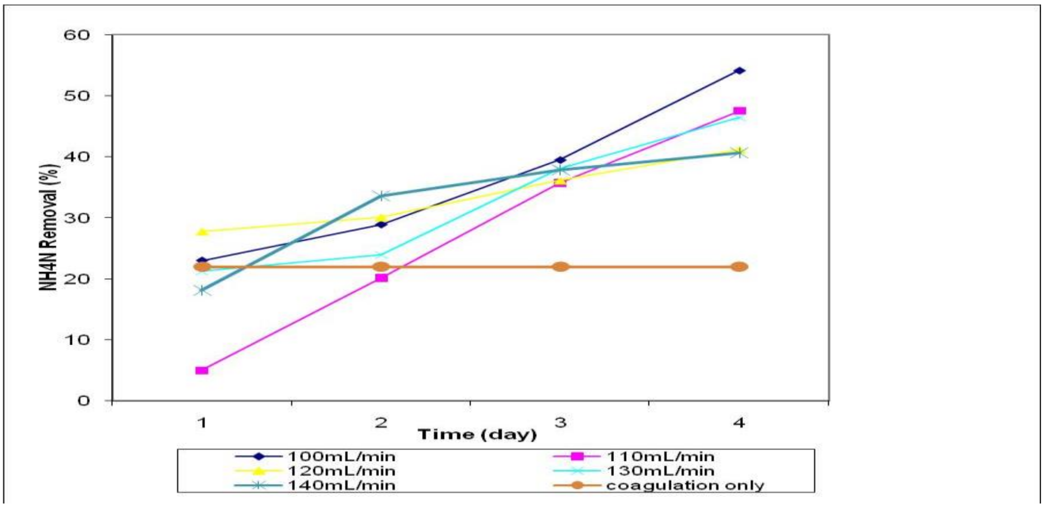

Figure 6. NH_4N Removal versus Time for Different Filtration Rates by using 5 µm PP and pH7.5

3.5 Different types of membrane effects

3.5.1 COD removal

The removal of COD was 40% under the filtration process. However, COD removal for the PP (1 - 5 µm) membranes was 64.8% and 59.4%, respectively, higher than 55% (Figure 5). No significant drop in the removal for these two membranes was noted. In the resin membrane, the percentage of COD removal dropped to 53.3%, and the removal for nylon membrane was less than 50%. Based on Figure (7), the smaller pore size exhibited a higher removal of COD. The percentage range of COD removal in the 1 µm membrane was 39.5%-64.8% compared with the 27.6%-59.4% removal in the 5 µm membrane.

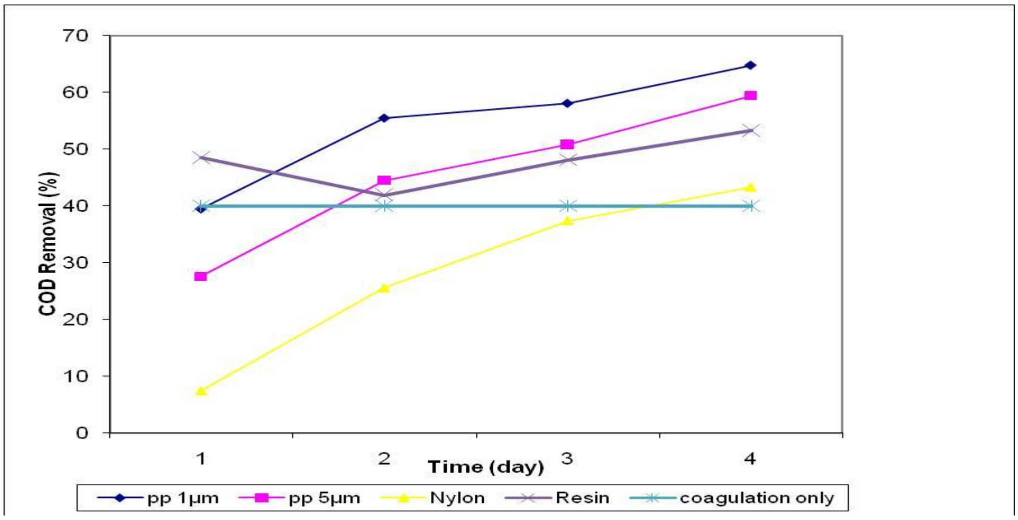

Figure 7. COD removal versus Time at a Constant Filtration Rate (100mL/min) and pH7.5 in Different Types of Membranes

3.5.2 Colour removal

The removal of colour was 75% prior to the filtration process. Moreover, 5 µm PP exhibited the highest colour removal at 85.3% and 1 µm PP had a removal rate of 85.2%. Colour removal using resin and nylon was 78.8% and 76.59%, respectively (Figure 8).

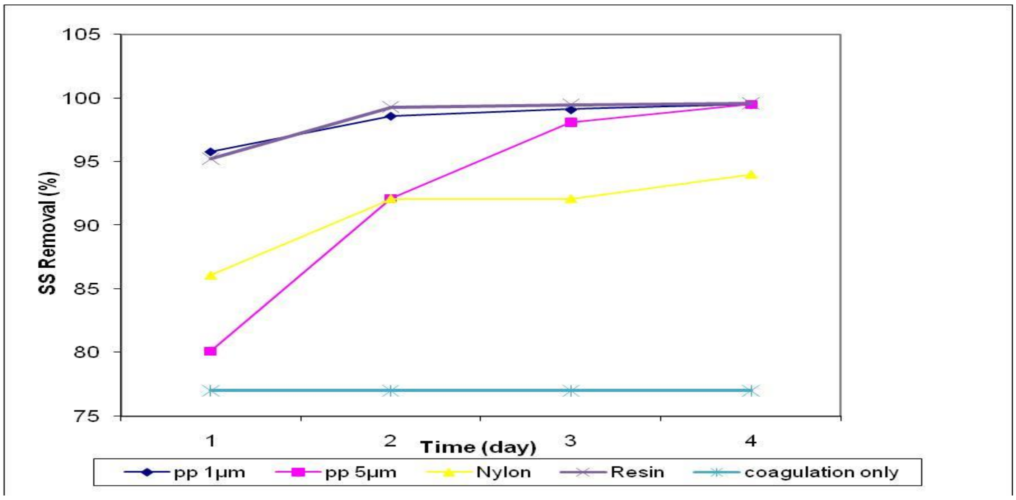

Figure 8. Colour Removal versus Time at a Constant Filtration Rate (100mL/min) and pH7.5 in Different Types of Membranes

3.5.3 Suspended Solids removal

Figure (9) shows the percentage of SS removal using different types of membrane filters at a constant filtration rate of 100 mL/min. SS removal of SS was 77% prior to the filtration process. However, SS removal was the lowest a 94% reduction after using the nylon membrane.

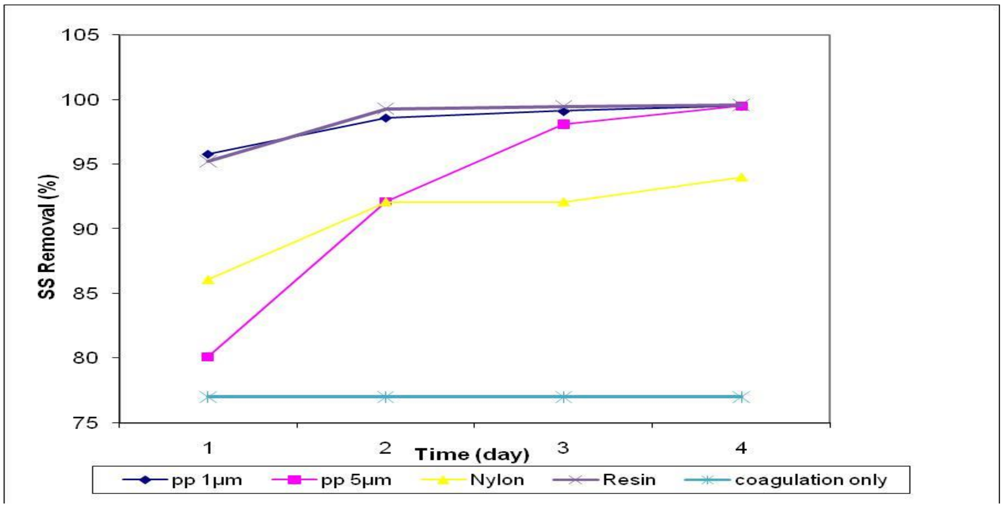

Figure 9. Suspended Solids Removal versus Time at a constant Filtration Rate (100mL/min) and pH7.5 in Different Types of Membranes

3.5.4 Ammoniacal Nitrogen (NH_4N) removal

Figure (10) shows the graph of (NH_4N) percentage removal using different types of membrane filters at a constant filtration rate of 100 mL/min. NH_4N removal was 22% prior to the filtration process. There was no significant difference in the removal of NH_4N using PP (1 and 5 µm) and nylon membranes. The removal for these membranes was in the ranged from of 8%-52%. The range of NH_4N removal was 75%-99% for the resin membrane.

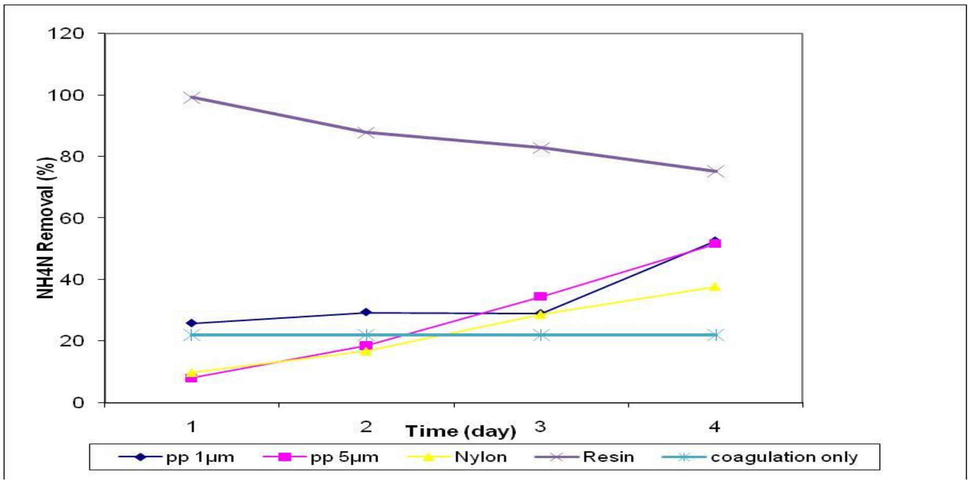

Figure 10. NH_4N Removal versus Time at a Constant Filtration Rate (100mL/min) and ph7.5 in Different Types of Membranes

3.5.5 One-way ANOVA on colour removal efficiency

Table (3) indicates that there is a significant difference in the COD, colour and SS, NH_4N removals among all the filtration rates. The *P* value was 0 with colour and SS removal. With regard to the colour, the best colour removal was highest at the filtration rate of 100mL/min (mean=86.06%) compared with the other filtration rates, such as 110mL/min (mean=85.88%), 120mL/min (mean=84.13%), 130mL/min (mean=78.0%) and 140mL/min (mean=82.8%). The best SS removal was highest at the filtration rate of 100mL/min (mean=98.1%) compared with the other filtration rates, such as 110mL/min (mean=96.78%), 120mL/min (mean=94.0%), 130mL/min (mean=90.05%) and 140mL/min (mean=96.88%). The *P* value for NH_4N was 0.556. The best removal was highest at the filtration rate of 100mL/min (mean=36.4%) compared with the filtration rates, such as 110mL/min (mean=27.11%), 120mL/min (mean=33.78%), 130mL/min (mean=32.41%), and 140mL/min (mean=32.58%). The *P* value of COD removal was 0.037. The best removal was achieved using PP (1 m) membrane with mean removal of 54.48% as compared with the others, PP (5m) (mean=45.58%), nylon (mean=28.45%) and resin (mean=47.95%).

Table 3. ANOVA results for COD, Colour, SS, and N_4HN Removals efficiency at different filtration rates using 5µm PP and pH7.5

Parameters	*F* -Value	Sig.
COD removal	3.378	.037 (S)
Colour removal	10.014	.000 (S)
SS removal	15.653	.000 (S)
N_4HN removal	18.813	.556 (NS)

4. Conclusion

Membrane filtration was applied for leachate treatment from the PBLS after a coagulation process using PAC in this study. The first part of this study was focused on the effect of various filtration rates in removing the parameters, namely, COD, colour, SS and NH_4N. The percentage of COD, colour, SS, and NH_4N removal after the coagulation process was lower than that of the removal after the coagulation and filtration processes at different filtration rates using 5 µm PP and pH 7.5. The percentage of COD, colour, SS, and NH_4N removal was increased at a low filtration rate. The results obtained would be better if the filtration rates decreased. Based on the experimental results, the filtration rate of 110 mL/min exhibited the highest performance in removing COD at 67.3%, and the filtration rate of 100 mL/min showed the highest performance in removing colour at 89.4%, SS at99.5%, and NH_4N at 54.2%. The filtration rate of 130 mL/min showed the poorest results for colour and SS removals and the filtration rate of 140 mL/min gave the poorest results for COD and NH_4N removals. This method is suitable only for leachate treatment of effluent from PBLS with a high concentration of SS. The second part of this study was focused on the effect of various membranes, namely, nylon, resin, and PP, with a pore size of 1 and 5 µm used for leachate treatment. The percentage of parameters removal after coagulation was lower than that after the coagulation and filtration processes at a constant filtration rate of 100 mL/min and pH 7.5 using different types of membrane filters. PP was the most efficient in parameters removal compared with the others. However, 1 µm PP was better than 5 m PP in removing COD at 64.8%, SS at 99.5% and colour at 85.18%. The 5µm PP membrane was the most efficient in NH_4N removal compared with the others, achieving 43.6% for colour removal. The nylon membrane gave the poorest results in removing the parameters, especially for NH_4N, colour, and COD removal at a range of 28%-35%. Overall, the 1 µm PP at a filtration rate of 100 mL/min exhibited the highest results in parameters removal.

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