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Treatment of Landfill Leachate by Crossflow Microfiltration and Ozonation

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ABSTRACT

Laboratory scale experiments were conducted for the treatment of landfill leachate using crossflow microfiltration (CFMF) with periodic backflush as a declogging technique. Powdered activated carbon (PAC) was used as for pretreatment, and ozonation was used as posttreatment for CFMF. Single channel tubular ceramic membranes of 0.2 and 1.2 μm pore size were used in this study. The results showed that permeate flux increases with an increased dose of PAC added to the leachate. When membranes of different pore sizes were compared, the 0.2-μm membrane performed better than the 1.2-μm membrane, giving a higher flux as well as higher removal of color and COD. The optimum PAC dose for CFMF was found to be 30 g/L whereas the optimum value for the batch test was found to be 60 g/L.

INTRODUCTION

In recent years, solid waste management and disposal has become one of the most important environmental concerns. Landfills have served for many decades as ultimate disposal sites for all types of wastes: residential, commercial, and industrial, both innocuous and hazardous (1). Leachate is the wastewater produced when water and other liquids seep through solid and liquid waste deposited in landfills. It is a complex wastewater containing high quantities of organic matter, color, heavy metals, and suspended solids, and it leads to oxygen depletion, increased hardness, metal precipitation, and increased toxicity, and it affects the complex aquatic food chain. In order to prevent ground water and surface water pollution, leachate must be collected and treated before discharge (2).

Leachate can be treated by biological, physical, or chemical methods or a combination of these methods. The treatment methods to be used depend upon the chemical composition of the leachate and on the age of the landfill. In the case of a young landfill, the leachate mainly contains low molecular weight organics which are best treated by biological processes. It is known that physicochemical processes are appropriate for leachate treatment when the leachate is low in volatile degradable organics, i.e., the BOD_5/COD ratio is less than 0.1 and/or the molecular weight of most of the organics is greater than 500 g/mol. Thus, physicochemical techniques are best used following biological treatment or for treating a leachate from a mature and stabilized landfill.

It is difficult to treat a leachate by a single method because of its complex nature which depends on the type of solid waste disposed, seasonal variations, etc. Most landfill leachate cannot be treated adequately by conventional physicochemical or biological processes. Microfiltration process with a membrane pore size of 0.02 to 10 μm removes discrete suspended solids and colloids which are not easily settled and has been applied for treatment of various types of wastewaters (3). Ozone is a common oxidizing agent used for the treatment of water and wastewater, mainly due to its lack of hazardous by-products. It has been found satisfactory for transforming high molecular weight compounds to low molecular weight compounds and thus increasing the biodegradability of organic substances (4).

The research described in this paper was aimed at developing a treatment method for landfill leachate such that the effluent from the treatment may be safely discharged to a biological treatment plant or, in some cases, to natural receiving water. To achieve this, a combination of techniques was used to treat the leachate.

MATERIALS AND METHODS

The leachate employed in this study was collected from the On-nooch solid waste disposal site, Bangkok. The characteristics of the leachate are given in Table 1.

The batch tests were conducted using a jar test apparatus to determine the treatability of PAC. For this, various doses of powdered activated carbon (PAC, from 0 to 90 g/L) were added to a 200-mL leachate sample

TABLE 1
Characteristics of the Leachate Used in this Study

Parameter	
pH	8.5
Conductivity (mmhos/cm)	14.95
Suspended solids (mg/L)	588
Total solids (mg/L)	12,500
Color (°Hazen)	15,000
COD (mg/L)	4,704
BOD (mg/L)	240
Cu (mg/L)	0.343
Pb (mg/L)	0.22
Mn (mg/L)	0.621
Cd (mg/L)	0.12
Fe (mg/L)	10.75
Zn (mg/L)	2.66

and mixed at 150 rpm for 60 minutes at room temperature and then filtered through Whatman GF/C filter paper to remove the PAC before analysis. The process evaluation was done on the basis of color and COD removal efficiency.

The leachate was treated by crossflow microfiltration using periodic backflush as the declogging technique. PAC was used to pretreat the leachate and ozonation as posttreatment.

Batch experiments were conducted with PAC and crossflow microfiltration (CFMF) for the treatment of the leachate. For this, various amounts of PAC (0 to 30 g/L) were added to 8 L of the initial leachate sample in a container and stirred with a mechanical stirrer for 30 minutes. This was then put into the storage tank of the CFMF unit and was passed directly through the membrane in the crossflow mode. All the experiments were conducted for 180 minutes.

Experiments were also conducted in which, after mixing PAC with raw leachate for 30 minutes, the mixture was allowed to settle for 30 minutes. Then only the supernatant was passed through CFMF unit to analyze the effect of solids settling on the permeate flux.

The laboratory-scale experimental setup for CFMF with periodic backflush as the declogging technique used in this study is shown in Fig. 1. After pretreatment with PAC the sample was pumped from the storage tank under pressure to the tubular membrane filter. The operating pressure was maintained at 85 kPa and the crossflow velocity was 3 m/s during all experimental runs. A small portion of permeate was allowed to accumulate

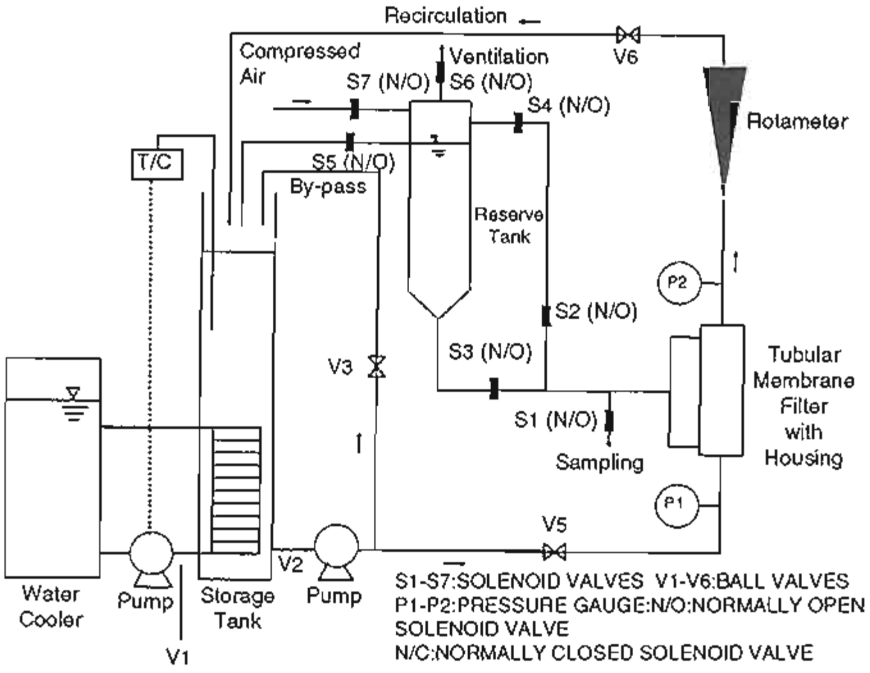


FIG. 1 Experimental setup of CFMF with periodic backflush cleaning technique.

in the reserve tank, and it was subsequently used for backflushing. The remaining portion of filtrate was collected in a container. The concentrate was circulated back to the storage tank, thus increasing the solid concentration in the storage tank. The temperature was maintained at 30 ± 2°C by an automatic temperature controller. The programmable controller together with the solenoid valves was employed for automatic operation and control of backflush duration (T_b = 1 second) and filtration time (T_f = 1 minute).

Single channel tubular ceramic membranes with pore sizes of 0.2 and 1.2 μm were used in this study. The membranes were Membralox ceramic microfilters from SCT (Society Ceramique Technique), France. The outer and inner diameters of these membrane are 10 and 7 mm, respectively; the total tube length of the membrane is 250 mm; the effective permeation area is 4550 mm^2; and the bursting pressure is greater than 30 bars.

Posttreatment was done by passing the ozone into the CFMF filtrate which had been pretreated with PAC. The ozone generator used for this study was a lab-scale module. Ozone was passed at a rate of 70 mg/L of oxygen (flow rate of oxygen = 2.5 mL/s) for 1 hour into a 1-L glass bottle reactor containing 750 mL filtrate. A magnetic stirrer was used for continuous mixing at constant speed. The ozonation time was kept constant for all experiments to enable comparison of the results. Samples were collected at 15 minute interval during ozonation and were analyzed for color and COD.

In order to study treatability by ozone alone, the ozone was contacted directly with the raw leachate. The other experimental conditions were kept the same for analysis of the effects of preozonation and postozonation after treatment of leachate with the membrane and PAC.

RESULTS AND DISCUSSION

The characteristics of the leachate used in this study are given in Table 1. From this table it is seen that the leachate is highly colored (15,000 °Hazen). The leachate has a low BOD_5/COD ratio (0.05–0.07), which shows that the organic matter in the leachate has low biodegradability. A low BOD_5/COD ratio, high pH, and low metal concentration indicate that the leachate is from a stabilized landfill which can best be treated by physicochemical processes (2).

The leachate also contains a lot of dissolved or colloidal matter, since the suspended solids are 588 mg/L while the total solids are 12,500 mg/L. This indicates that the colloidal matter (<1 μm) is 11,912 mg/L.

Batch experiments using a jar test apparatus were conducted to analyze the effect of the amount of PAC on color and COD removal of the leachate. Figure 2 depicts the effect of PAC dose on the treatment of leachate. It can clearly be seen that as the amount of PAC increases, the percent removal of color and COD also increases. From doses of 5 to 60 g/L there is a rapid increase in the removal of color (from 25 to 94.7%) and COD (from 20.5 to 88.8%). But when the PAC dose is increased from 60 to 90 g/L, the rate of color and COD removal decreases. Thus, 60 g/L may be considered to be the optimum dose.

Treatment by Powdered Activated Carbon and Crossflow Microfiltration

The effect of PAC dose on permeate flux was studied by using different doses of PAC (0 to 30 g/L) for two membranes having pore sizes of 0.2 and 1.2 μm. The other operating conditions were kept constant.

Effect on Permeate Flux

The permeate flux was monitored regularly at 15 minute intervals. Figure 3 shows the effect of different doses of PAC on permeate flux using a 0.2-μm membrane.

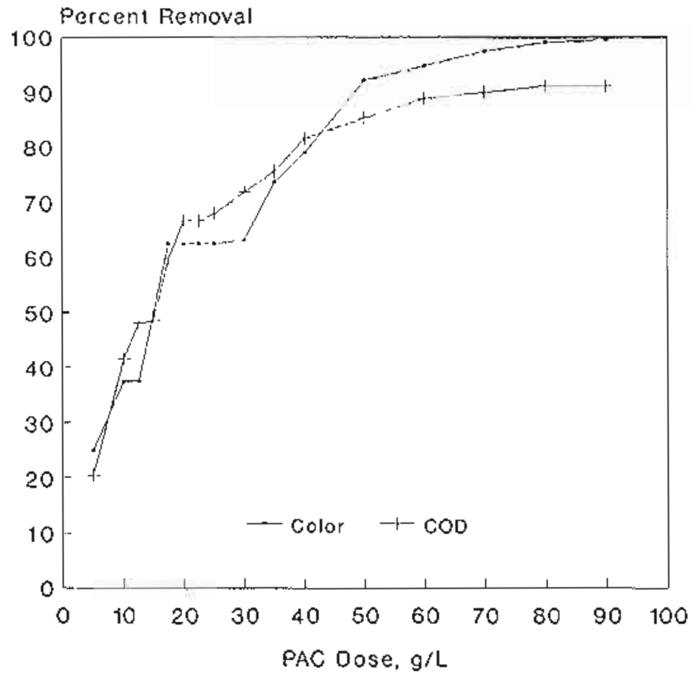


FIG. 2 Effect of PAC dose on treatment of leachate.

When raw leachate was passed through the membrane with no PAC added, a reduction in flux from 153 to 96 L/m^2·h in 3 hours was observed, indicating that the colloids deposited in the form of a thin film on the surface of membrane, thereby causing membrane clogging. When PAC was mixed prior to filtration, the drop in the permeate flux with respect to time was less (Table 2 and Fig. 2) and a more or less stable flux was obtained from the start of the experiment.

When a PAC dose of 20 g/L was added, the increase in permeate flux was 59.3% in comparison to the flux obtained with raw leachate. Similarly, a PAC dose of 30 g/L produced a 68.7% higher flux (Table 2). This comparison of flux was made at the end of the experiment, i.e., after 180 minutes operation of the CFMF unit. This clearly reflects that the addition of PAC produces increased flux.

Figure 3 and Table 2 clearly indicate that as the quantity of PAC is increased, the permeate flux increases although the suspended solids also increase with increasing dose. This is mainly due to the fact when more PAC is added, colloids are adsorbed on the PAC surface. Because the

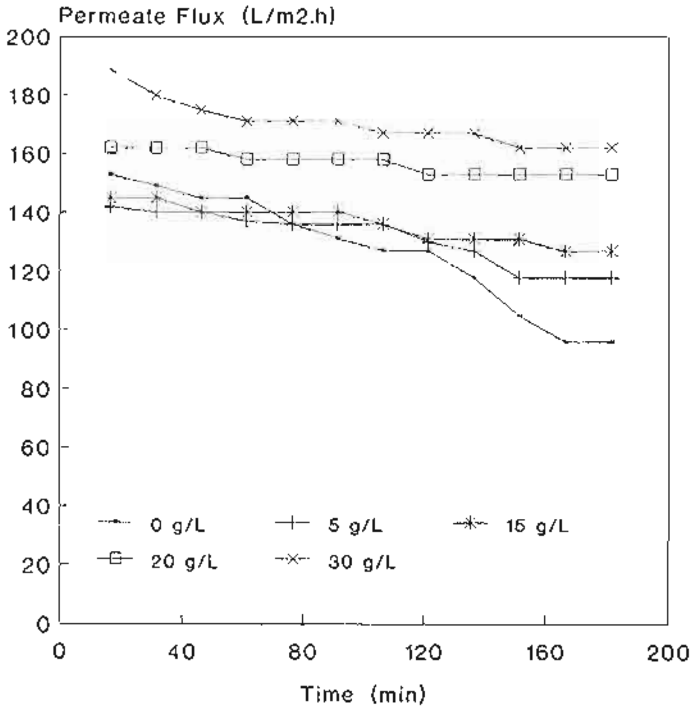


FIG. 3 Variation of permeate flux with time at different doses of PAC using a 0.2-μm membrane.

TABLE 2
Percent Increase in Permeate Flux with PAC Dose and Decrease in Flux with Time (0.2 μm Membrane)

PAC dose (g/L)	Permeate flux ($L/m^2 \cdot h$)		Percent flux increase due to PAC[b]	Percent flux reduction in 3 hours
	Start[a]	End[a]		
0	153	96	—	37.2
5	142	118	22.9	16.9
15	145	127	32.2	12.4
20	162	153	59.3	5.5
30	189	162	68.7	14.2

[a] "Start" means at 15 minutes and "End" means at 180 minutes of CFMF operation.

[b] Calculated with fluxes at the end of the experiment and on the basis of flux obtained with a 0-g/L PAC dose.

size of these PAC particles is much larger than the pore size, they cause only external membrane fouling and are flushed away with an applied crossflow velocity of 3 m/s. In addition, the PAC acts as an abrasive and keeps the membrane surface clean by scouring the deposited filter cake, and thus helps to increase the flux.

The effect of PAC dose on permeate flux was also studied by using a 1.2-μm membrane. Figure 4 shows the effect of PAC dose on permeate flux. Note that the permeate flux increases with PAC dose, as also shown in Table 3. The percent reduction in flux decreased as the PAC dose increases. The increase in flux at a dose of 30 g/L was 56.3% when compared to the flux obtained with raw leachate.

Although the flux increases with PAC dose for both the 0.2- and 1.2-μm membranes, the permeate flux obtained using the 1.2-μm membrane (Table 3) was always less than the flux from the 0.2-μm membrane (Table 2) for the same dose of PAC. This is also illustrated in Fig. 5. For example,

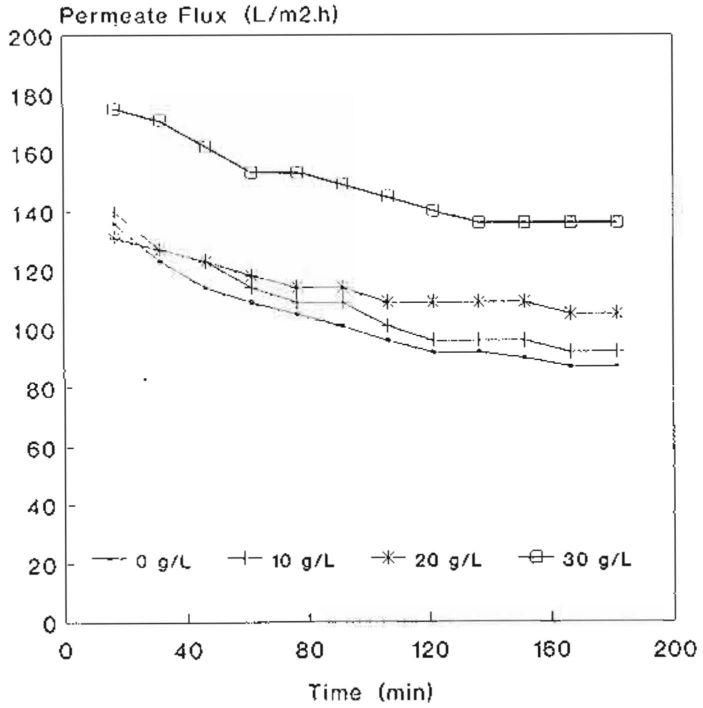


FIG. 4 Variation of permeate flux with time at different doses of PAC using a 1.2-μm membrane.

TABLE 3
Percent Increase in Permeate Flux with PAC Dose and Percent Decrease in Flux with Time (1.2 μm Membrane)

PAC dose (g/L)	Permeate flux (L/m²·h)		Percent flux increase due to PAC[b]	Percent flux reduction in 3 hours
	Start[a]	End[a]		
0	136	87	—	36.0
10	140	92	5.7	34.0
20	131	105	20.6	19.8
30	175	136	56.3	22.2

[a] "Start" means at 15 minutes and "End" means at 180 minutes of CFMF operation.

[b] Calculated with fluxes at the end of the experiment and on the basis of flux obtained with a 0-g/L PAC dose.

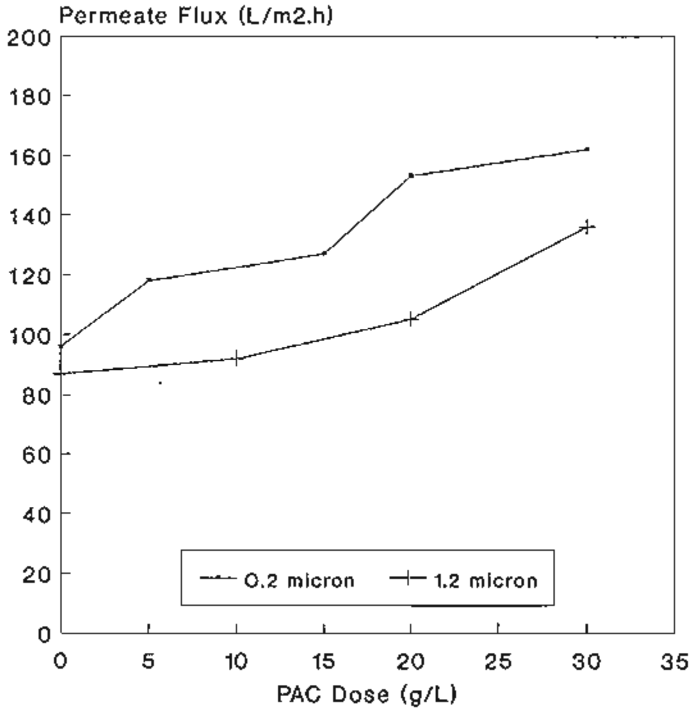


FIG. 5 Comparison of permeate flux at different doses of PAC using 0.2 and 1.2-μm membranes.

when the raw leachate without the addition of PAC was filtered through the 0.2-μm membrane, a flux of 96 L/m^2·h was obtained whereas the larger pore-size membrane (1.2-μm) gave a lesser flux of 87 L/m^2·h. This may be due to the presence of a large amount of dissolved organics and colloids (<1 μm) in the leachate or because a portion of the PAC particles which enter the pores of the 1.2-μm membrane cause internal fouling of the membrane and cannot be effectively removed by the backflush technique. Thus, a better performance was obtained by using the 0.2-μm membrane compared to using the 1.2-μm membrane.

Effect of Settled PAC on Permeate Flux

To find the effect of settling after adsorption on permeate flux, the leachate was mixed with different doses of PAC and allowed to settle for 30 minutes and the supernatant was passed through the membrane in the crossflow mode. The membrane with a pore size of 0.2-μm was used for this study. The results have been compared with those obtained when the leachate and PAC mixture was passed through the membrane without allowing the PAC to settle. Figure 6 shows the effect of PAC dose on PAC settling.

The suspended solids concentration in the supernatant after settling was 5.45, 5.72, and 6.95 g/L in comparison to 12.63, 16.91, and 19.62 g/L for a PAC dose of 15, 20, and 30 g/L respectively, when PAC was not allowed to settle.

Figure 6 show that the flux obtained with settling is always lower than the flux obtained without settling for all the PAC doses considered. Normally, an increase in suspended solids concentration should decrease the flux. This contradiction can be explained by the fact that after settling, some unsettleable colloids which are not adsorbed by PAC cause internal clogging of the pores as well as the creation of a slime colloidal deposition on the external surface of the membrane. This slime cannot be eliminated by an applied crossflow velocity of 3 m/s. The increase in flux with increased suspended solids due to higher doses of PAC is also attributed to the fact that the PAC acts as an abrasive and helps to remove the cake deposited on the membrane surface.

These results suggest that for a membrane with a 0.2-μm pore size used in combination with PAC for the treatment of leachate, settling is not desirable.

Effect of PAC Dose on Color and COD Removal in Combination with CFMF

The results obtained for color and COD at different doses of PAC using a 0.2-μm filter are shown in Fig. 7. When raw leachate was filtered, 76.6%

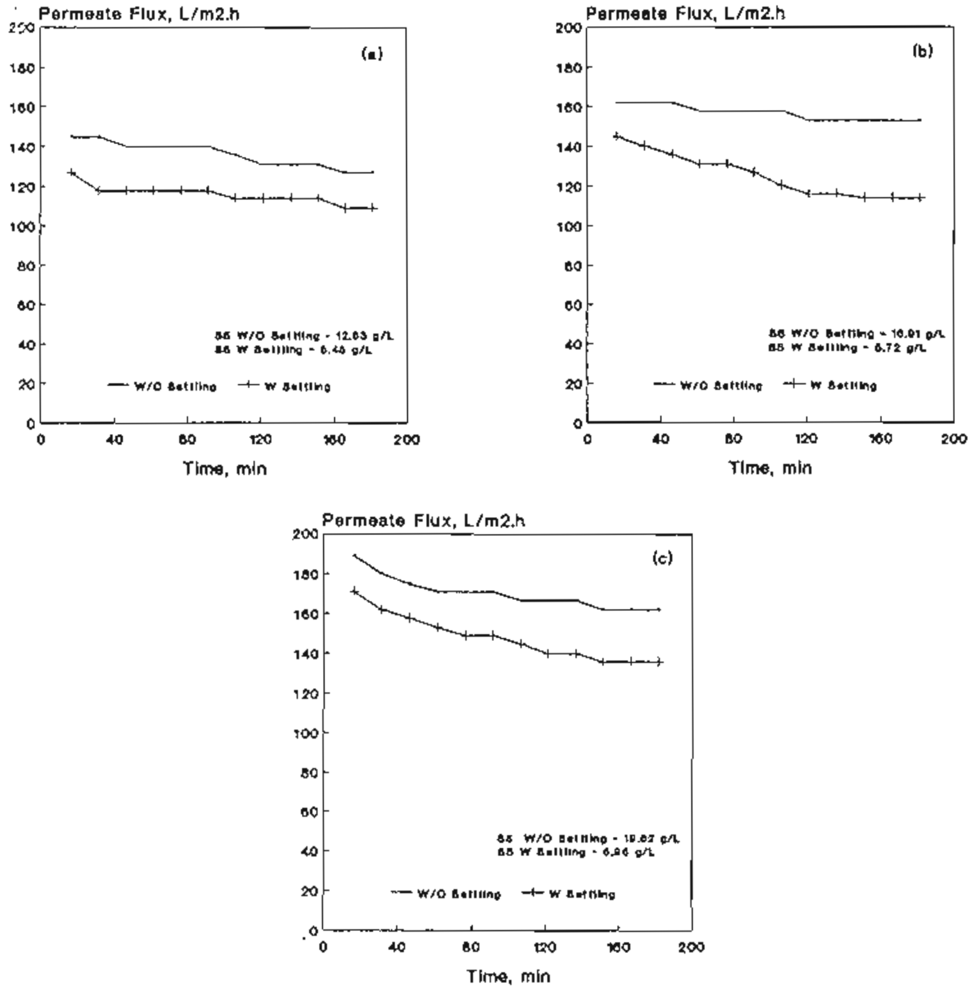


FIG. 6 Effect on permeate flux using a 0.2-μm membrane with settling and nonsettling of PAC: (a) 15 g/L, (b) 20 g/L, (c) 30 g/L.

of the color and 62.3% of the COD were removed from the initial color (15,000 °Hazen) and the COD (4838 mg/L), respectively. This indicates that the membrane itself is capable of reducing the color and COD to a large extent. The increased color and COD removal with the addition of PAC is mainly due to the increased adsorption of suspended solids and colloids. When a PAC dose of 30 g/L was added to the leachate, 96% of the color and 89.1% of the COD were removed.

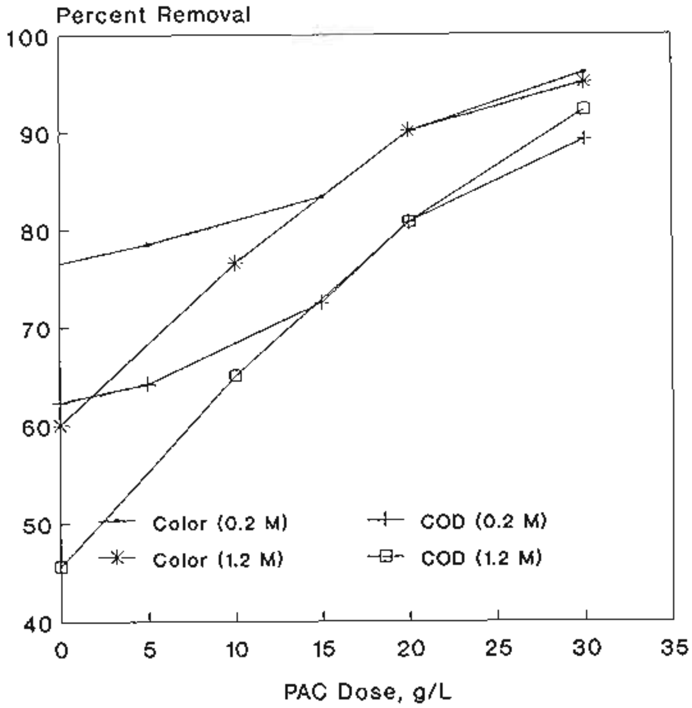


FIG. 7 Effect of PAC dose on color and COD of filtrate obtained from CFMF using 0.2 and 1.2 μm membranes.

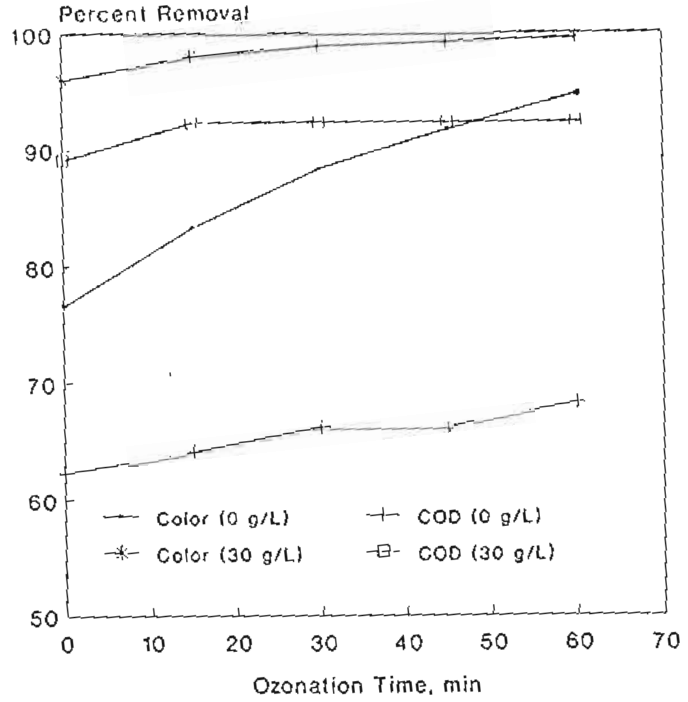
The results for color and COD obtained with a 1.2-μm filter at various doses of PAC are depicted in Fig. 7. For the treatment of raw leachate with this membrane, a removal of 60% of the color and 45.6% of the COD was observed. With the addition of PAC, there was more removal of color and COD.

When the results obtained with 0.2 and 1.2 μm membranes are compared, both membranes show a similar trend of increasing the percent removal of color and COD with increasing PAC dose. When raw leachate was filtered, a higher removal of color and COD was observed with the 0.2-μm membrane than with the 1.2-μm membrane due to higher rejection of colloids and suspended solids because of the difference in the pore sizes.

It is interesting to note (Fig. 7) that for PAC doses of 15 g/L and larger, the removal of color and COD is nearly the same for both membranes. With a PAC dose of 15 g/L or less, the 0.2-μm membrane gives better results. When the flux obtained from these membranes is considered, the 0.2-μm membrane gives a higher flux for all doses.

Ozonation of CFMF Filtrate Pretreated with PAC

The results of filtrate ozonation obtained from a 0.2-μm membrane with a PAC dose of 0 and 30 g/L are depicted in Fig. 8 and Table 4. From Table 4 it is seen that there is an increase in color removal with an increase in time of ozonation of the filtrate obtained from CFMF with an increase in PAC dose. Not much change in COD was observed. When filtered raw leachate was ozonated for 60 minutes, a color removal of 94.6% and a COD removal of 68.2% were observed. When the color of the filtrate is high (which is the case when no or a small quantity of PAC is added), the ozone is effective in reducing the color. The effluent standard of color of 500 °Hazen can be met after less than 1 hour ozonation when PAC is added.



i. 8 Effect of ozonation on color and COD removal of CFMF filtrate using a 0.2-μm membrane with PAC doses of 0 and 30 g/L.

TABLE 4
Percent Removal of Color and COD by Individual and Combination of Techniques (0.2 μm Membrane)

PAC dose (g/L)	PAC		PAC + CFMF		PAC + CFMF + O_3		CFMF		Ozone	
	Color	COD	Color	COD	Color	COD	Color	COD	Color	COD
1	2	3	4	5	6	7	4–2	5–3	6–4	7–5
0	0.0	0.0	76.6	62.3	94.6	68.2	76.6	62.3	18.0	5.9
5[a]	20.0	26.8	80.0	64.2	96.6	69.6	60.0	37.4	16.6	5.4
15[b]	46.6	52.8	83.3	72.5	96.6	73.3	36.7	19.7	13.3	0.8
20	53.3	60.5	90.0	80.7	99.0	84.6	36.7	20.2	9.0	3.9
30	66.6	67.6	96.0	89.1	99.5	92.3	29.4	21.5	3.5	3.2

[a] Ozonation for 50 minutes.

[b] Ozonation for 40 minutes.

A similar trend of color and COD removal was observed with the 1.2-μm membrane as with the 0.2-μm membrane.

From the above results it is evident that ozone helps in removing the color but that not much COD is reduced. The latter may be due to the presence of refractory organics. Partial oxidation by ozone helps in the conversion of refractory organic compounds to biodegradable organics or a shift toward low molecular weight organics which can be further degraded by biological processes.

Table 4 gives the percent removal of color and COD of leachate by PAC, PAC + CFMF (with a 0.2-μm membrane), and PAC + CFMF + ozone. Figure 9 gives graphical presentations of the effectiveness of each process in removing color and COD.

From Table 4 it can be seen that the 0.2-μm membrane removes 76.6% of the color and 62.3% of the COD when raw leachate is filtered, but the permeate flux is lower compared to when PAC is added to the leachate.

TABLE 5
Amount of PAC Required by Individual and Combined Techniques to Attain the Same Removal Efficiency

Technique (s)	Percent removal: Color	Percent removal: COD
PAC (60 g/L)	94.7	88.8
PAC (30 g/L) + CFMF	96.0	89.1
PAC (5 g/L) + CFMF + O_3	96.6	69.6

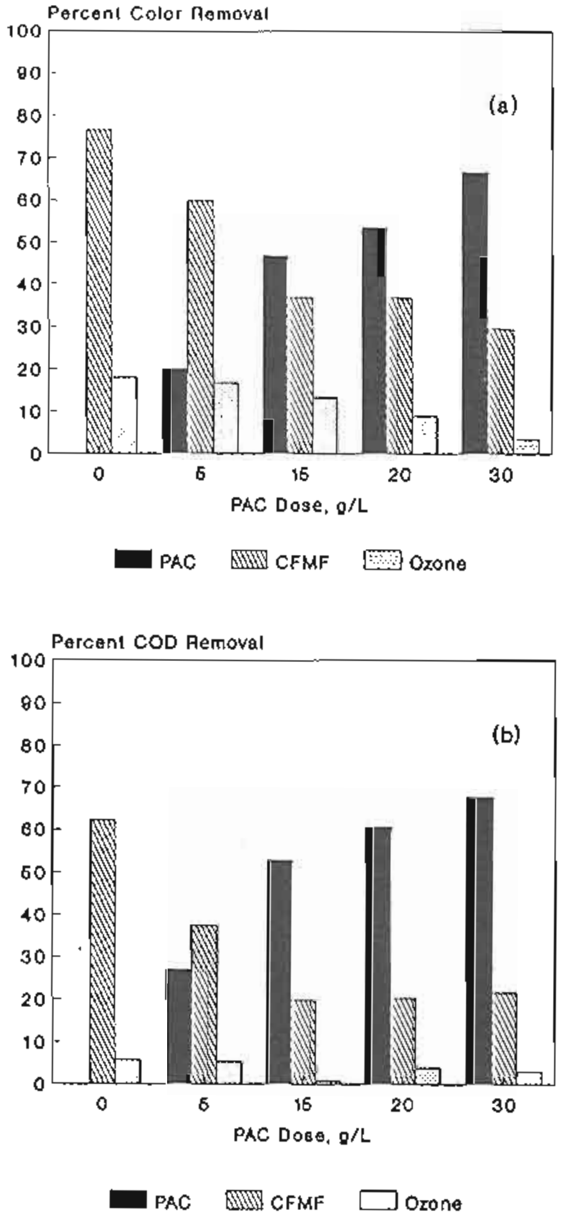


FIG. 9 Removal efficiency of PAC, CFMF, and ozone: (a) color, (b) COD. Membrane pore size: 0.2 μm.

There is an increase trend of permeate flux and color and COD removal with the dose of PAC. So a combination of PAC and CFMF is more suitable than an individual process in order to have a higher flux and at the same time a better removal efficiency.

Table 5 compares the quantity of PAC required in the treatment of leachate by PAC alone and in combination with CFMF and ozone to achieve almost the same level of color and COD removal.

It can be seen from Table 5 that to achieve a removal efficiency of 94.7% of the color and 88.8% of the COD by PAC alone, a dose of 60 g/L is required. The same and even slightly more (96% of the color and 89.1% of the COD) removal was obtained with a half dose of PAC (30 g/L) when this was coupled with crossflow microfiltration. Only 5 g/L of PAC is enough to obtain a color removal of 96.6% when this dose is used with CFMF and ozone with an ozone rate 10.5 mg/min and an ozonation period of 60 minutes. This clearly indicates that using a combination of PAC, CFMF, and ozone can reduce the amount of PAC required and thereby reduce the sludge produced.

Table 6 provides the percent removal of color and COD of leachate by PAC, PAC + CFMF (with a 1.2-μm membrane) and PAC + CFMF + ozone. These results agree with those obtained with the 0.2-μm membrane.

Treatment of Raw Leachate by Ozone

To study the feasibility of using only ozone for the treatment of leachate, the ozone was passed directly into the raw leachate at a rate of 10.5 mg/min for a 700-mL sample. A graphical representation is given in Fig. 10. Color and COD removal increases with the time of ozonation. As seen from the figure, a color removal of about 90% was achieved in 240 minutes

TABLE 6
Percent Removal of Color and COD by Individual and Combination of Techniques (1.2 μm Membrane)

PAC dose (g/L)	PAC		PAC + CFMF		PAC + CFMF + O_3		CFMF		Ozone	
	Color	COD	Color	COD	Color	COD	Color	COD	Color	COD
1	2	3	4	5	6	7	4–2	5–3	6–4	7–5
0	0.0	0.0	60.0	45.6	83.3	53.4	60.0	45.6	23.3	7.8
10	33.3	46.5	76.6	65.0	94.6	74.5	43.3	18.5	18.0	9.5
20	53.3	60.5	90.0	80.7	99.0	88.4	36.7	20.2	9.0	7.7
30	66.6	67.6	95.0	92.1	99.5	93.7	28.4	24.5	4.5	1.6

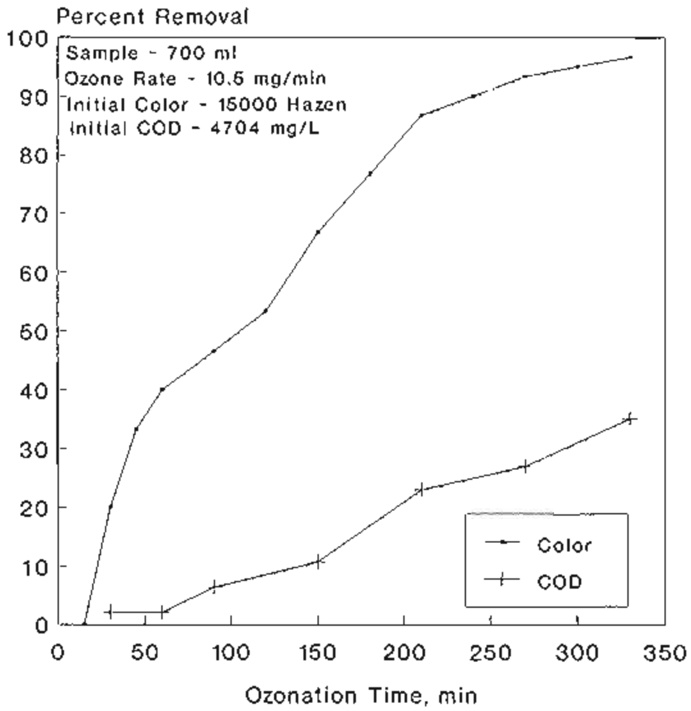


FIG. 10 Effect of ozonation on color and COD removal of raw leachate.

for a 700-mL sample. The corresponding COD removal was only about 25%, which suggests that only the color of such a complex leachate can be removed but that a long ozonation time is required. It can be concluded that for the removal of pollutants, using only ozone may not be the best choice due to its high cost, unless the COD of the wastewater is very low.

CONCLUSIONS

A more or less stable and increased flux is obtained with the augmentation of the PAC dosage for both the types of membrane (0.2 and 1.2 μm), but a higher flux was obtained with a 0.2-μm membrane compared to a 1.2-μm membrane with and without the addition of PAC. The flux obtained with a 0.2-μm membrane after settling was lower compared to the flux obtained without settling for all doses of PAC added to the leachate, which indicates that settling is not required when a mixture of PAC and leachate is treated with CFMF.

When PAC alone is used to treat the leachate, a dose of 60 g/L is required to achieve a removal of 94.7% of the color and 88.8% of the COD, but when PAC and CFMF are coupled together, only 30 g/L of PAC is required to achieve the same level of removal. A further reduction in PAC requirement was observed when this treatment was coupled with ozonation. This clearly indicates that a combination of PAC plus CFMF (using a 0.2-μm membrane) plus ozone is a better choice for treating the leachate because it not only reduces the amount of PAC required, but also produces less sludge.

Ozone was found to be effective in reducing the color, but not much of an effect on COD removal was observed. Direct ozonation of raw leachate reduced the color to 500 °Hazen but required a long time of ozonation; the corresponding COD removal was very low. This suggests that direct ozonation may not be the best choice to treat the leachate.

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