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Removal of non-biodegradable compounds from stabilized leachate using VSEPRO membrane filtration

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Abstract

In this paper, the technical applicability of the vibratory shear-enhanced processing reverse osmosis (VSEPRO) membrane system for the treatment of stabilized leachate from the NENT landfill in Hong Kong was studied using both 99 and 96% NaCl rejection membranes. The performance of VSEPRO for NH_3-N and COD removal was evaluated and compared to the performances of other individual and/or combined treatments from other studies. Without any pH adjustment, it was found that the rejection rates of VSEPRO were 96% for COD and 98% for NH_3-N with initial COD and/or NH_3-N concentrations of 8000 and 2620 mg/L, respectively. It was evident that pH adjustment of raw leachate to acidic conditions (pH 6.0) slightly improved the rejection rates for COD (97%) and NH_3-N (99.6%). These results were satisfactory compared to other physicochemical treatments. It is important to note that the treatment of leachate using VSEPRO was able to meet the local effluent limit for COD of lower than 200 mg/L and for NH_3-N of less than 5 mg/L. This result suggests that VSEPRO is technically applicable and appealing for the treatment of stabilized leachate.

Keywords: Landfill leachate management; Municipal solid waste; Recalcitrant compounds; Waste treatment

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1. Introduction

With a total population of more than seven millions in 2004 [1], Hong Kong generates a huge amount of municipal solid waste (MSW) every day. About 11,030 tonnes of MSW and 6730 tonnes of construction and demolition waste are disposed off in landfills daily [2]. Presently, there are three on-going landfills in Hong Kong (SENT, NENT and WENT) with the overall capacities of 37×10^6, 39×10^6 and $61 \times 10^6\,m^3$, respectively [3].

With an increasing age, the solid waste landfilled undergoes various physicochemical and biological changes. As a result, the degradation of the organic fraction of the waste in combination with the percolating rainwater leads to the generation of a highly contaminated wastewater called "leachate". The leachate contains a high strength of ammoniacal-nitrogen (NH_3-N) and recalcitrant compounds (as indicated by its COD value) [4]. Because the leachate may find its way into the underlying groundwater, the generation of landfill leachate poses potential long-term hazards to the surrounding environment.

Of the toxic compounds present in the landfill leachate, NH_3-N has been identified as one of the major toxicants to aquatic organisms, as confirmed by toxicity analyses using organisms such as *Salmo gairdnieri* [5]. With a concentration of higher than 100 mg/L, untreated NH_3-N can stimulate algal growth, inhibit nitrification and have toxic effects on living organisms [6].

Although the characteristics of landfill leachate depend on the type of MSW, the degree of solid waste stabilization, the hydrology of the site, the age of the landfill and the stage of MSW decomposition [7], raw leachate from local landfills are commonly characterized by high strengths of NH_3-N (2500–5000 mg/L) and COD (5000–20,000 mg/L) and a low ratio of BOD_5/COD (less than 0.1) [8]. Therefore, unless properly treated, the leachate that seeps from a landfill and enters the groundwater would pose potentially serious hazards to the surrounding environment.

In Hong Kong, landfill leachate has been commonly treated on-site using biologically aerobic reactors. Although this practice can reduce the loading of the reactors, it requires a large installation space and high costs (US\$ 13/m^3 of treated effluent) [9]. Moreover, the results of this treatment are sometimes not cost-effective. Therefore, there is a growing need to find other alternatives to improve the treatment performance of leachate.

Reverse osmosis (RO) has been employed for the treatment of landfill leachate in Yachiyo (Japan) [10]. Using vibratory shear-enhanced processing (VSEP), the application of RO offers advantages such as high rejection rates and reduced space needs. Due to its shearing force, VSEPRO can treat landfill leachate containing recalcitrant compounds of varying strengths and compositions with limited membrane fouling [11]. RO may enable the treated effluent to meet the effluent limit of discharge standards imposed under local environmental legislation.

In this paper, the technical applicability of the vibratory shear-enhanced processing reverse osmosis (VSEPRO) membrane system for leachate treatment from the North East New Territories (NENT) landfill in Hong Kong was studied using 99 and 96% NaCl rejection membranes. The effects of pH on its treatment performance were investigated. The treatment performance of VSEPRO for COD and/or NH_3-N removal from the leachate was evaluated and compared to the performance of other individual and/or combined treatments from other studies.

2. Materials and methods

Raw leachate was collected from the NENT landfill (Hong Kong) and stored at 4°C before subsequent use. The leachate was immediately characterized according to the Standard Methods

Table 1
List of equipment

Parameter	Equipment used
pH	pH meter model Orion 710A
Concentration of NH_3-N and/or COD	Spectrophotometer model Spectronic 20 Gendys
Conductivity	Conductivitimeter type YSI 63
Total organic carbon (TOC)	TOC analyzer type Shimadzu 5000A

[12] for the following parameters: pH, COD, BOD_5, NH_3-N, alkalinity (as $CaCO_3$), turbidity, total organic carbon (TOC) and conductivity. Adjustment of pH was carried out using 0.1 N NaOH and/or 0.1 N HCl. All equipments employed in this study are presented in Table 1.

2.1. Membrane selection

About 50 L of leachate was fed to a VSEPRO unit type L/P (Fig. 1). The unit consisted of a driving system that generated vibration, a membrane module, a torsion spring that transferred vibration to the membrane module and a system for vibration. It vibrated the membrane with 60 Hz of frequency. Unless otherwise mentioned, leachate treatment was carried out at ambient temperature (25–27°C) and at 29 bar of pressure for the overall studies. Two membranes with 99 and 96% NaCl rejection and 0.5 ft^2 of area were evaluated. The membrane with high permeates and stable flux flow rate was selected for further study.

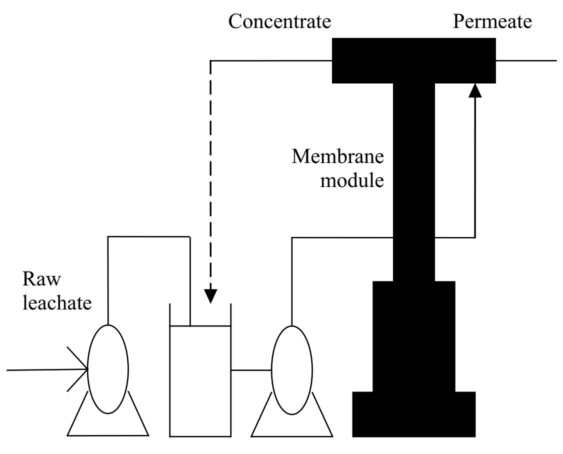

Fig. 1. Scheme of VSEPRO unit.

2.2. Treatment of leachate using VSEPRO

Without any pH adjustment, 50 L of raw leachate were fed to a VSEPRO unit for treatment. The first permeate was collected and about 45 L leachate from the first permeate were recirculated to the unit for further treatment. The treated effluent from the first and second permeates was analyzed for COD, NH_3-N, BOD and NO_3^--N. To minimize the escape of NH_3 gas to the atmosphere, the pH of raw leachate was adjusted to 6.0. The same treatment using VSEPRO was also employed for raw leachate with pH adjustment [13].

2.3. Chemical analysis for NH_3-N and/or COD

The changes in NH_3-N and/or COD levels were determined colorimetrically according to the Standard Method [12]. An indophenol-blue color was generated in the reaction between NH_3-N in the sample and coloring reagents. Absorbances were measured at a wavelength (λ) of 626 nm for NH_3-N and 600 nm for COD analyses. The removal efficiency of NH_3-N and/or COD (E) after VSEPRO treatment was defined as $E(\%) = [(C_0 - C_e)/C_0] \times 100$, where C_0 and C_e are the initial and equilibrium concentrations of NH_3-N and/or COD in the leachate (mg/L), respectively.

2.4. Statistical analysis

To ensure the reproducibility of the collected data, experiments were carried out at least in triplicate and the mean value of the data sets is presented. If the relative error exceeded 1.0%, the data would be disregarded and a fourth experiment conducted until the error fell within an acceptable range. A paired *t*-test was performed to evaluate the data for any significant difference in terms of COD and/or NH_3-N removal before and after the pH adjustment of leachate. The test undertaken after the Kolmogorov–Smirnov test confirmed the normality of variable distribution ($p > 0.05$). The results were analyzed using SPSS 11.00 Windows version with a confidence interval of 95% ($p \leq 0.05$) [14].

3. Results and discussion

3.1. Leachate characteristics from the NENT landfill

The characteristics of the raw leachate sample from the landfill are presented in Table 2.

It is found that the leachate has a low BOD_5/COD ratio of 0.09, typical of raw leachate from the methanogenic phase. This suggests that most of the organic materials have been converted to methane, thus decreasing the ratio of biodegradability of the leachate. The low ratio of BOD_5/COD indicates that the raw leachate was from a stabilized landfill, which is better treated with physicochemical techniques such as RO than with biological process. In addition to organic compounds, the leachate might contain inorganic substances such as heavy metals, as indicated by the conductivity value. It is important to note that the VSEPRO membrane system has the ability to remove not only organic but also inorganic compounds, with high rejection rates for heavy metals.

Table 2
Characteristics of the leachate

pH	8.01
COD (mg/L)	8000
BOD_5 (mg/L)	700
BOD_5/COD	0.09
NH_3-N (mg/L)	2620
NO_3^--N (mg/L)	290
Alkalinity (as $CaCO_3$) (mg/L)	12,986
Turbidity (NTU)	3900
TOC (mg/L)	2590
Conductivity (mmS/cm)	12.5

Table 3
Rejection performances of 99 and 96% NaCl rejection membranes

Type of RO membrane	% NaCl rejection			Flux (GFD)
	NH_3-N	COD	NO_3^--N	
99% NaCl rejection	96.0	98.7	100.0	38.4
96% NaCl rejection	86.0	85.4	99.9	24.5

3.2. Membrane selection

In a preliminary study, the RO membrane of 99% NaCl rejection was found to have a relatively higher flux (38.4 GFD) than that of the 96% NaCl rejection (24.5 GFD) (Table 3). The former also had slightly higher rejection rates (COD: 98.7%, NH_3-N: 96%, NO_3^--N: 100%) than the latter (COD: 85.4%, NH_3-N: 86%, NO_3^--N: 99.9%) (*t*-test; $p > 0.05$). These results concluded that the 99% NaCl rejection membrane was more suitable than the 96% NaCl rejection membrane for the treatment of landfill leachate.

3.3. Effect of pH on the treatment performance of VSEPRO

Without any pH adjustment, the removal of NH_3-N from raw leachate increased slightly from 83 (the first permeate) to 98% (the second permeate) with an initial concentration of 2620 mg/L (Table 4). At the same 80% permeate recovery, a high rejection was also obtained for the removal of COD (96%), BOD (91%) and NO_3^--N (99.6%) with initial concentrations as listed in Table 2. VSEPRO treatment could also improve the biodegradability of the leachate, as indicated by the increasing BOD_5/COD ratio

Table 4
Rejection of vibratory shear-enhanced processing reverse osmosis (VSEPRO) (with 80% permeate recovery) for raw leachate

	NO_3^--N	BOD_5	COD	NO_3^--N	Ratio of BOD_5/COD
Rejection of the 1st permeate (%)	82.7	91.1	86.5	99.6	0.24
Overall % rejection	98.0	91.1	95.9	99.6	

from 0.09 (before treatment) to 0.24 (after treatment). These results suggested that most large organic compounds were degraded into smaller compounds during treatment [15].

After pH adjustment to 6.0, no significant improvement of removal performance was observed for COD and NH_3-N (paired *t*-test; $p \leq 0.05$). The NH_3-N removal improved slightly from 98% to an almost complete removal (99.6%) at the same initial concentration of 2620 mg/L (Table 5). This phenomenon could be attributed to the fact that the pH adjustment of leachate to acidic conditions (pH 6.0) shifted the equilibrium of ammonia from the gas to the liquid phase, resulting in a higher removal of NH_3-N from the leachate, as shown in Eq. (1):

$$NH_{3\,(g)} + H^+_{(aq)} \Leftrightarrow NH^+_{4(aq)} \qquad (1)$$

For the cost-effectiveness of treatment, further investigation is required to find out if NH_3-N concentration in the leachate should be removed by adjustment to pH 11 for ammonium stripping treatment, or by struvite precipitation if the remaining concentration is still high. With a stoi-chiometric ratio of $Mg{:}NH_4{:}PO_4 = 1{:}1{:}1.3$, struvite precipitation was employed to remove NH_3-N from leachate [16]. The precipitation of struvite forms an insoluble compound which can be easily separated from the liquid, as shown in Eq. (2):

$$MgCl_2{\cdot}6H_2O + Na_2HPO_4 + NH_4^+ \Leftrightarrow MgNH_4PO_4{\cdot}6H_2O\downarrow + 2\,NaCl \qquad (2)$$
$$pKs = 12.6\ (25^{\circ}C)$$

It was reported that struvite precipitation could remove 90% NH_3-N with an initial concentration of 2240 mg/L. This result indicated that the NH_3-N removal by VSEPRO was still higher than that by struvite precipitation, although the precipitation could convert NH_3-N into a valuable nitrogen fertilizer such as urea.

At pH 6.0, it was also found that the COD removal increased slightly from 96 to 97% at the same initial concentration of 8000 mg/L (Table 5). This could be attributed to the change of surface charges in the RO membrane and organic compounds such as humic substances in the leachate. With the decreasing pH, the surface charge of the membrane became less negative, while humic substances were more protonated, thus becoming more hydrophilic. As a result, the electrostatic attraction between the humic substances and the membrane surface would be enhanced, causing a higher COD removal from the leachate [7].

It is important to note also that the treatment of stabilized leachate using VSEPRO was able to meet the local effluent limit in Hong Kong for COD of lower than 200 mg/L and for NH_3-N of less than 5 mg/L. These results suggest that

Table 5
Rejection of vibratory shear-enhanced processing reverse osmosis (VSEPRO) (with 80% permeate recovery) for leachate with pH adjustment

	NH_3-N	BOD_5	COD	NO_3^--N
Rejection of the1st permeate (%)	91.7	93.5	89.8	99.8
Overall rejection (%)	99.6	93.5	97.0	99.8

VSEPRO is technically applicable and appealing. Moreover, no biological process is required for further treatment of the leachate, thus reducing possible operational costs.

Although landfill leachate varies in strengths and compositions, the application of VSEPRO for leachate treatment offers many advantages compared to other treatment techniques such as an upflow anaerobic sludge blanket (UASB) and chemical precipitation. VSEPRO has the ability to remove NH_3-N and recalcitrant organic compounds with high rejection rates and prevent foulants from accumulating on the membrane surface using a vibratory drive mechanism [17].

Table 6
Comparison of COD and NH_3-N removal from stabilized leachate using various treatments

Type of treatment	Type of Precipitant/ adsorbent/ membrane	Dose (g/L)	Initial concentration in leachate (mg/L)		Pressure (bar)	BOD/ COD	pH	Removal efficiency/ rejection rate (%)		References
			COD	NH_3-N				COD	NH_3-N	
Individual treatment										
Ammonia stripping	$Ca(OH)_2$	11	5850	3260	–	0.60	11.0	–	94	[18]
Precipitation	Struvite	NA	7511	5618	–	0.22	8.5–9.0	–	98	[4]
RO	SW30-2521	–	3840	NA	52	0.31	6.0	98	NA	[19]
NF	NA	NA	17,000	3350	NA	0.03	6.4	96	NA	[20]
Adsorption	PAC	6	5690	2215	–	NA	NA	95	NA	[21]
VSEPRO	–	–	8000	2620	29	0.24	8	96	98	Present study
	–	–	8000	2620	29	0.24	6	97	100	Present study
Combined treatments										
RO + UASB	–	–	35,000	1600	NA	–	7.4	99	99	[22]
RO + activated sludge	–	–	6440	1153	NA	0.70	NA	99	99	[23]
Nanofiltration + adsorption + ozonation	Desal 5K GAC O_3	– NA NA	4000	NA	8.5	NA	6.5	99	NA	[24]
Ultrafiltration + adsorption	GAC	NA	3050	NA	–	0.55	7.0	97	NA	[25]
Ozonation + adsorption	O_3 GAC	5	4970	700	–	0.17	8–9	90	NA	[15]
Coagulation + fenton oxidation	$FeCl_3$ Fe(II)/ H_2O_2	0.5 NA	7400	NA	–	0.06	8.5	90	NA	[26]
RO + evaporation	AD	–	19,900	30	60	0.20	6.4	88	97	[27]

Remarks: *NA: Not available.

This mechanism allows the membrane to maintain a higher and constant permeate rate, enabling it to process larger volumes of leachate at a lower cost. Moreover, VSEPRO can eliminate membrane fouling with a stable flux in operation and it needs less installation space and a shorter treatment time, thus improving its cost-effectiveness.

3.4. Comparison of removal performance with other physicochemical techniques for leachate treatment

To evaluate the performances of different treatments investigated in the present study, a comparative study is presented in terms of pH, dose required (g/L) and initial concentration of COD and NH_3-N (mg/L) in the leachate. Table 6 summarizes the COD and/or NH_3-N removal efficiency of various individual and/or combined treatments in previous studies. Although it has a relative meaning due to the different testing conditions such as pH, temperature, the strength of wastewater, seasonal climate and hydrology site, this comparison is useful to evaluate the overall treatment performance of each technique for a decision-making process.

Struvite precipitation has achieved 98% treatment efficiency for NH_3-N removal with an initial concentration of 7511 mg/L. An outstanding COD removal of 96–98% has also been demonstrated by nanofiltration (NF) and reverse osmosis (RO) with COD concentrations ranging from 3840–17,000 mg/L. Among the various combined treatments, the combination of RO and UASB (with initial COD and NH_3-N concentrations of 35,000 and 1600 mg/L, respectively) has achieved an almost complete removal of COD and NH_3-N. When compared to the treatment efficiencies presented in Table 6, the removal of COD (97%) and/or NH_3-N (99.6%) by VSEPRO treatment (with pH adjustment) in this study is very satisfactory. Moreover, the total COD and NH_3-N removal in this study was higher (COD: 97% out of 8000 mg/L; NH_3-N: 99.6% out of 2620 mg/L) than that of another study by using the combined treatment of RO and activated sludge [23], which removed 99% of COD with an initial concentration of 6440 mg/L and 99% of NH_3-N with an initial concentration of 1153 mg/L.

4. Conclusions

It is evident from the study that the use of VSEPRO for the removal of COD and/or NH_3-N from landfill leachate is technically applicable and appealing. The advantages of using this membrane system for the treatment of leachate are the simplicity of the treatment and its ability to treat stabilized landfill leachate of varying strengths and compositions. Without any pH adjustment, it was found that the rejection rates of VSEPRO were 96% for COD and 98% for NH_3-N with initial COD and/or NH_3-N concentrations of 8000 and 2620 mg/L, respectively. pH adjustment of the raw leachate to acidic conditions (pH 6.0) did not significantly improve the rejection rates for COD (97%) and NH_3-N (99.6%). These results were very satisfactory compared to those of other physicochemical treatments. It is important to note that the treatment of leachate using VSEPRO could meet the local effluent limit for COD of lower than 200 mg/L and for NH_3-N of less than 5 mg/L. This result suggests that VSEPRO is technically applicable and appealing for the treatment of stabilized leachate. No biological process is required for further treatment of the leachate, thus reducing operational costs.

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