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Research Paper

Assessing Membrane Performance for Landfill Leachate Treatment in Accordance with Local Regulatory Requirements

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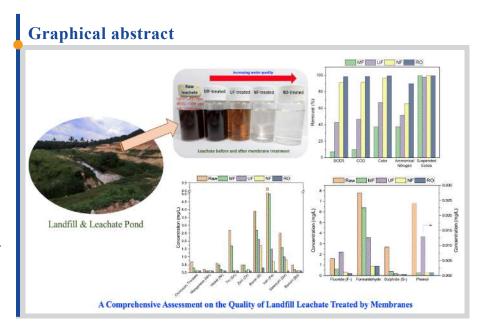
Regulations COD

BOD

Metal ions

Highlights

- First comprehensive assessment on the quality of local leachate treated by membranes
- NF and RO membranes showed better quality of treated leachate than microporous membranes
- RO membrane could meet most of the local discharge standards
- A pretreatment is required to improve BOD and ammoniacal nitrogen removal of the membrane



Abstract

Poor landfill management is always associated with environmental problems such as the discharge of leachate that does not comply with environmental regulations. In this work, we carried out a comprehensive assessment of the quality of leachate treated by four different types of commercial polymeric membranes according to the local environmental quality regulations. Our findings revealed that although the microfiltration (MF) and ultrafiltration (UF) membranes were able to exhibit significantly higher water permeability compared to the reverse osmosis (RO) and nanofiltration (NF) membranes during leachate treatment, the properties of the treated landfill leachate in terms of biochemical oxygen demand (BOD₅), chemical oxygen demand (COD), color and ammoniacal nitrogen (AN) is far below the water treated by the NF/RO membranes. Furthermore, the NF/RO membranes were able to achieve a lower concentration of metallic elements as well as other parameters such as fluoride and sulfide in their permeates compared to the loose MF/UF membranes. Owing to the smallest pore size of the RO membrane, the quality of the permeate produced was the best among the membranes tested. However, a proper pretreatment process is still required before the RO membrane in order to comply with the stringent regulations of BOD₅ (≤ 20 mg/L) and AN (≤ 5 mg/L).

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1. Introduction

Statistics reveal that Malaysia currently has 165 landfills, eight sanitary landfills, and three inert landfills for materials such as concrete and sand [1]. On average, Malaysia produces >30,000 tons of municipal solid waste daily with food waste accounting for the largest fraction.

Owing to the increasing population and rapid urbanization in Malaysia, solid waste management has become more and more challenging [2]. A study showed that approximately 95% of municipal solid waste is disposed of in landfills while the rest is used for composted activity and resource recovery

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[3]. According to a study conducted by Abunama et al. [4], it is estimated that for every ton of waste disposed in landfills, annual leachate generation is found to be 148 L and 79 L for humid and semi-arid landfills, respectively. In Europe, a report estimated that between 200 L and 1000 L of leachate could be produced per ton of waste landfilled each year [5]. Based on the daily municipal solid waste produced in Malaysia (30,000 tons), it is estimated that >4.44 million liters of leachate are produced annually.

Landfill leachate is a liquid that forms when rainwater or other liquids come into contact with wastes stored in a landfill [6]. As the liquid passes through the wastes, it picks up various contaminants including organic and inorganic compounds, heavy metals, and pathogens, making it a significant environmental concern if it is not properly collected and treated [7]. The traditional industrial leachate treatment methods can be categorized into three major groups, (a) leachate transfer, i.e., recycling and integrated treatment with sewage, (b) physical and chemical methods such as oxidation, adsorption. chemical precipitation, coagulation/flocculation, sedimentation and (c) biological process, e.g., aerobic and anaerobic treatment [8]. Although these methods have been practiced for years, the tightening discharge standards in many countries coupled with the aging of landfill sites (with more and more stabilized leachates) have caused the conventional treatments insufficient to achieve the desirable outcomes [9,10].

One of the emerging treatment methods that has been considered to address the limitations of the traditional leachate treatment methods is membrane technology which involves the use of semipermeable film to separate contaminants from the leachate [11-13]. This method offers a promising alternative that could improve the separation efficiency of leachate treatment as well as reduce the system footprint, leading to reduced environmental impact of leachate. Currently, membrane bioreactor (MBR) which combines conventional biological treatment process with microporous membrane technology is the most widely used membrane technology for landfill leachate treatment [14-17]. Overall, the MBR could offer several unique advantages such as consistent effluent quality, reduced sludge production, and high removal rates against suspended solids and organic matter, but it also has some limitations including ineffective to remove heavy metal ions and some chemicals present in leachate. A large number of MBR studies only focused on the removal efficiencies of membranes against several key parameters such as total suspended solids (TSS), chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), total nitrogen (TN), color and phosphorus [16,18-20]. Determining the efficiency of the membranes against these parameters is not sufficient considering the stringent regulation of leachate discharge. In Malaysia, for instance, the discharge of leachate must comply with a total of 29 parameters including heavy metal ions and some critical chemical compounds such as cyanide, formaldehyde, and phenol [21].

In view of this, the main objective of this work is to carry out a comprehensive assessment of the quality of leachate treated by membrane technology according to the local environmental quality regulations. Four different classes of commercial polymeric membranes with different pore sizes ranging from microfiltration (MF) to reverse osmosis (RO) will be evaluated for their respective performance in removing various pollutants from the leachate collected from a local landfill. To the best of our knowledge, no study has been carried out to investigate the performance of different membrane properties for the treatment of leachate by assessing not only the typical parameters such as TSS, BOD₅, COD, and color but also other parameters as required by the regulations.

2. Experimental

2.1. Leachate Collection

The leachate sample was collected from a landfill located in Johor on January 5, 2023. No pretreatment (i.e., sieving) was performed on the leachate sample on the site. The sample was stored in the water container and kept in a cooler bag packed with ice packs during transportation. After reaching the laboratory, the water samples were kept at 4°C in a refrigerator before it was used for experiment the following days. The leachate exhibits significant characteristics, as outlined in Table 1. Notably, it demonstrates elevated concentrations of BOD5, COD, ammoniacal nitrogen, and suspended solids. Furthermore, the local leachate displays relatively substantial levels of various heavy metals, namely iron, boron, zinc, and selenium. Additionally, relatively high levels of chemicals such as formaldehyde and sulfide have been detected.

Table 1
Properties of the leachate collected from the local landfill and its comparison with the literature data and the acceptable conditions for discharge in Malaysia.

Parameter	^a Value	Literature (min- max) (Roy et al., 2018) [22]	^b Environmental Quality Regulations in Malaysia	
Temperature (°C)	24	n/a	40	
pH	8.42	3.0-9.2	6.0-9.0	
BOD ₅ at 20°C	2100	30-72,000	20	
COD	3000	81-185,000	400	
Color (ADMI)	213	n/a	100	
Ammoniacal Nitrogen as N	3395	1.3-21,180	5	
Suspended Solids (SS)	1300	90-33,700	50	
Mercury (Hg)	< 0.001	n/a	0.005	
Cadmium (Cd)	< 0.01	0-1.58	0.01	
Chromium Hexavalent (Cr(VI))	< 0.01	n/a	0.05	
Chromium Trivalent (Cr(III))	0.7	n/a	0.20	
Arsenic (As)	< 0.01	n/a	0.05	
Lead (Pb)	< 0.1	0-0.52	0.10	
Copper (Cu)	0.1	0.01-2.34	0.20	
Manganese (Mn)	0.2	n/a	0.20	
Nickel (Ni)	0.6	0-4.43	0.20	
Tin (Sn)	2.7	n/a	0.20	
Zinc (Zn)	0.5	0.01-37.5	2.0	
Boron (B)	3.9	n/a	1.0	
Iron (Fe)	9.2	n/a	5.0	
Silver (Ag)	< 0.1	n/a	0.10	
Selenium (Se)	2.5	n/a	0.02	
Barium (Ba)	0.5	n/a	1.0	
Cyanide (CN ⁻)	< 0.01	n/a	0.05	
Fluoride (F ⁻)	1.6	n/a	2.0	
Formaldehyde	7.8	n/a	1.0	
Phenol (C ₆ H ₅ OH)	0.024	n/a	0.001	
Sulphide (S ⁻)	2.7	n/a	0.05	

^a All the parameters are in the unit of mg/L (equivalent to ppm), except for the temperature, pH, and color.

 Table 2

 Properties of flat sheet polymeric membranes used in this work.

Membrane	Model/Manufacturer	Membrane material/structure	Pore size/ MWCO
Microfiltration	Porafil/Macherey-Nagel	Polytetrafluoroethylene/Asymmetric	0.45 micron
Ultrafiltration	PS20/RisingSun Membrane Technology (Beijing) Co. Ltd.	Polysulfone/Asymmetric	20,000 Dalton
Nanofiltration	NF3/RisingSun Membrane Technology (Beijing) Co. Ltd.	Polyamide/Thin film composite	100-400 Dalton
Reverse osmosis	RO5/RisingSun Membrane Technology (Beijing) Co. Ltd.	Polyamide/Thin film composite	<1 nm

b Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Second Schedule: Acceptable Conditions for Discharge of Leachate.

2.2. Properties of Commercial Membranes

Four commercial flat sheet membranes obtained from different manufacturers were used in this work for leachate treatment. These membranes have different surface properties and are categorized as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) depending on their pore size, as shown in Table 2. The MF membrane has the largest pore size while the RO membrane has the smallest pore dimension.

2.3. Filtration Experiment and Water Analysis

Prior to the landfill leachate treatment at the laboratory, the polymeric membranes were respectively evaluated for their pure water permeability (L/m².h.bar) and salt rejection (%) using the commercial 316 stainless steel stirred cell (HP4750) manufactured by Sterlitech Corporation, USA. The filtration process was carried out in a dead-end mode using a feed solution of 300 mL. The working pressure for MF, UF, NF, and RO membranes was fixed at 1, 2, 10, and 15 bar, respectively for both pure water and salty solution filtration experiments.

The water permeability, J_{ν} (L/m²·h·bar) of the respective polymeric membrane can be obtained using Equation (1).

$$Jv = \frac{V}{A \times t \times P} \tag{1}$$

where V is the volume of permeate (L), A is the effective surface area of the membrane (m^2) , t is the duration required to collect the permeate (h) and P is the working pressure (bar). Salt rejection, R_S (%) was calculated using Equation (2).

$$R_{S} = \left| \frac{C_{initial} - C_{final}}{C_{initial}} \right| \times 100 \tag{2}$$

where $C_{\it initial}$ and $C_{\it final}$ are the concentration (mg/L) of NaCl in the feed and

permeate, respectively. The conductivity of the water sample was determined using a benchtop conductivity meter (4510, Jenway). The conductivity of the sample was then converted to concentration using a conductivity-concentration calibration curve.

For the landfill leachate treatment, the treated sample was collected from the membrane filtration process over a period of several hours from several identical stirred cells (HP4750, Sterlitech Corporation, United States). The permeate produced from the membrane was returned to the stirred cells from time to time and only a small amount of the permeate sample was collected for analysis. The quality of the permeate in terms of color (ADMI) and pH was determined in the laboratory using a UV-vis spectrophotometer (DR5000, Hach, United States) and a basic pH meter (pH5+, Eutech, United States), respectively. Further analysis of the quality of the treated samples was carried out by sending the samples to an accredited laboratory - Allied Chemists Laboratory Sdn Bhd (Johor Bahru, Malaysia). Table 3 presents the methods that were used to determine the value of each parameter. The quality of the treated samples was further compared with Malaysia's Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009. This regulation provides the acceptable conditions of 29 parameters for the discharge of leachate.

2.4. Characterization

Fourier transform Infrared spectroscope (FTIR, Nicolet iS10, Thermo Fisher Scientific, United States) was employed to compare the surface chemistry of respective membranes before and after the leachate treatment. This analysis was performed in an attenuated total-reflectance (ATR) mode to identify the functional groups of the membrane surfaces at wavenumber between 500 and 4000 cm⁻¹. Before the FTIR analysis, the membrane samples with dimensions of 20 mm \times 20 mm were treated in an oven at 40°C for 2 hours to remove any moisture from the membranes. An average of 32 scans was then conducted on the samples to yield the spectrum. A comparison of the leachate samples treated by membranes and the membranes used for the treatment process was also carried out by taking the respective photograph using a mobile phone (Galaxy S22+, Samsung, South Korea) with a built-in digital camera (12 MP ultra-wide camera). In addition, a scanning electron microscope (SEM, HITACHI TM3000) was utilized to compare the surface morphology of the membrane before and after leachate treatment. Energy dispersive X-ray (EDX) analysis based on JEOL's JSM-IT300LV was also performed on the fouled membrane samples to evaluate the chemistry of the foulants deposited on the membrane surface.

Table 3

The methods that were employed to analyze the quality of leachate samples treated by different membranes.

Parameters	^a Method					
General						
BOD ₅ at 20°C	APHA 5210 B / APHA 4500 O C					
COD	APHA 5220 C					
Color	HACH Method 10048					
Ammoniacal Nitrogen as N	APHA 2540 D					
Suspended Solids (SS)	APHA 4500-NH3 B&C					
	<u>Metals</u>					
Mercury (Hg)	APHA 3500 Hg / APHA 3112 B					
Cadmium (Cd)	APHA 3030 F / USEPA 6010 B					
Chromium Hexavalent (Cr(VI))	APHA 3500-Cr B					
Chromium Trivalent (Cr(III))	APHA 3030 F / USEPA 6010 B					
Arsenic (As)	APHA 3030 F / USEPA 6010 B					
Lead (Pb)	APHA 3030 F / USEPA 6010 B					
Copper (Cu)	APHA 3030 F / USEPA 6010 B					
Manganese (Mn)	APHA 3030 F / USEPA 6010 B					
Nickel (Ni)	APHA 3030 F / USEPA 6010 B					
Tin (Sn)	APHA 3030 F / USEPA 6010 B					
Zinc (Zn)	APHA 3030 F / USEPA 6010 B					
Boron (B)	APHA 3030 F / USEPA 6010 B					
Iron (Fe)	APHA 3030 F / USEPA 6010 B					
Silver (Ag)	APHA 3030 F / USEPA 6010 B					
Selenium (Se)	APHA 3030 F / USEPA 6010 B					
Barium (Ba)	APHA 3030 F / USEPA 6010 B					
	Others					
Cyanide (CN-)	HACH Method 8027					
Fluoride (F ⁻)	APHA 4500 F-D					
Formaldehyde	HACH Method 8110					
Phenol (C ₆ H ₅ OH)	HACH Method 8047					
Sulphide (S ⁻)	HACH Method 8131					

^aAPHA = American Public Health Association; USEPA = United States Environmental Protection Agency.

3. Results and Discussion

3.1. Water permeability and NaCl rejection

Fig. 1 compares the water permeability and NaCl rejection of four polymeric membranes selected in this work. With respect to pure water permeability, the MF membrane exhibits the highest value followed by UF, NF, and RO membranes. The decreasing trend is expected owing to the reduced membrane surface pore size. The water permeability of the membrane is inversely proportional to the solute rejection in which the higher the water permeability the lower the NaCl rejection and vice versa. When the membrane pore size is as small as the NF and RO membrane, the rejection rate of the membrane against NaCl is very high (>90%). However, it must be pointed out that the pure water permeability and rejection of each membrane might vary compared to the same class of membrane reported elsewhere [23-25]. This is due to many factors such as differences in average surface pore dimension, pore size distribution, surface roughness, and hydrophilicity of membrane as well as the use of different materials in manufacturing the membrane. For instance, the MF membrane exhibits only a marginal increase in pure water permeability compared to the UF membrane employed in this study. This modest improvement can largely be attributed to the hydrophobic properties of the MF membrane, composed of polytetrafluoroethylene (PTFE). This inherent hydrophobicity on the membrane surface results in considerable hindrance to water transport, particularly at low operating pressures (1 bar), leading to lower water permeability when compared to conventional MF membranes.

Compared to the pure water permeability, the leachate permeability of the membranes is significantly lower. This can be due to the presence of many solutes and dissolved ions in the leachate which contribute to the increased concentration polarization on the membrane surface. In addition, it is also possible that some of the solutes might cause membrane surface fouling which negatively affects the water permeability. As shown, the leachate permeability of the membranes decreases with decreasing the membrane pore size, i.e., MF (14.30 L/m².h.bar) > UF (5.62 L/m².h.bar) > NF (0.63 L/m².h.bar) > RO (0.13 L/m².h.bar).

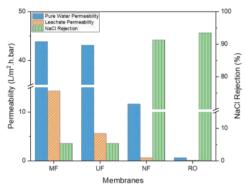


Fig. 1. Pure water permeability, leachate permeability, and NaCl rejection of different membranes.

3.2. Landfill Leachate Treatment

As shown in Fig. 2, it is very clear to see that the properties of the treated landfill leachate are gradually improved by using the membranes with decreasing pore size. Compared to the dark color of the raw leachate, the color intensity of the treated leachate is found to be significantly reduced when dense membranes (i.e., NF and RO) are used for the treatment. The RO-treated sample, in particular, is crystal clear, indicating the excellent quality of the permeate.



Fig. 2. Photographs of the leachate before and after polymeric membrane treatment.

Fig. 3 compares the FTIR spectrum of the respective polymer membrane before and after the landfill leachate treatment. As can be seen, the peak intensities of the UF, NF, and RO membranes at a broad region of 3000-3500 cm⁻¹ are decreased after the leachate treatment, indicating the presence of foulants on the membrane surface that covers the hydroxyl groups (-OH) of membranes. Since the MF membrane is made of polytetrafluoroethylene ((C₂F₄)_n), it does not show any characteristic peak at the wavenumber of 3000-3500 cm⁻¹. The peaks that appeared at 1148 and 1201 cm⁻¹ corresponded to the C-F stretching of the fluoro compound of the polytetrafluoroethylene. However, it must be noted that the membrane's spectrum at 3000-3500 cm⁻¹ is slightly altered after the leachate filtration, suggesting the existence of foulants on the membrane surface. Furthermore, several additional peaks are detected on the used MF membrane at 1660 and 1545 cm⁻¹. These peaks are possibly due to the alkene (C=C) and nitro compound (-NO) functional groups contributed by severe foulants deposition. Compared to other membranes, the surface of the MF membrane is obviously stained with more foulants (see Fig. 4(a)) and this could be possibly due to its largest pore size that makes the suspended solids easily trapped within its

Compared to the NF and RO membranes, it is found that the intensities of several peaks of the UF membrane are reduced to a greater extent. This can be explained by the fact that the pore dimension of the UF membrane is significantly larger than those of RO and NF membranes and this makes the membrane prone to the higher fouling propensity. Some characteristic peaks of the UF membrane (made of main polymer – polysulfone) that are altered after the leachate treatment are 3200–3550 cm⁻¹ (C–OH), 2900–3000 cm⁻¹ (CH), 1656 cm⁻¹ (H-O-H) and 688–871 cm⁻¹ (C=C). With respect to the NF and RO membrane, the characteristic peaks of the polyamide selective layer could be found at 1585, 1610, and 1660 cm⁻¹. These peaks are corresponded to the amide II, aromatic ring, and amide I, respectively. Since piperazine (PIP) is employed as the sole amine monomer to fabricate the NF membrane, the peak found at 1540 cm⁻¹ in the RO membrane is not able to be detected in the NF membrane. This is owing to the missing N—H bond in the amide formed with acid chloride (—RCON—) [261].

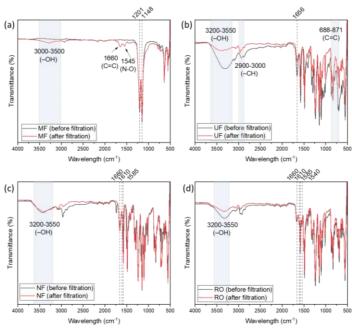


Fig. 3. FTIR spectrum of membranes before and after the leachate treatment, (a) MF membrane, (b) UF membrane, (c) NF membrane, and (d) RO membrane.

Fig. 4(b) compares the surface morphology of the membranes before and after leachate treatment. It is obvious that MF and UF membranes experience significant change on their surface, where the deposition of organic and inorganic foulants can be clearly observed. The EDX surface mapping conducted on the used membranes focused on analyzing the presence of

specific metal ions, namely Sn, B, and Fe that exhibit relatively higher concentrations in the leachate (as detailed in Table 1). The analysis also encompassed elements inherent to the polymeric structure of the membranes. In Fig. 4(c), the data illustrates the occurrence of Sn element exclusively on the loose membranes, namely MF and UF. Notably, this particular element is

conspicuously absent on the dense NF and RO membranes. Conversely, both B and Fe elements were not detectable on the surface of these membranes. A plausible explanation for their absence is attributed to the likelihood that they are not associated or bound with organic foulants similar to Sn. This distinction in the distribution of elements, with Sn being present primarily on the loose membranes while B and Fe remain undetected, suggests differing interactions or affinities of these elements within the membrane systems.

Fig. 5 provides a comprehensive assessment of the quality of landfill leachate treated by four different types of polymeric membranes. All the data are sorted out based on three categories, i.e., general parameters, metal-based parameters, and other parameters for ease of comparison. With respect to the general parameters (Fig. 5(a)), the trend is very clear, i.e., the smaller the membrane pore dimension the higher the rejection rates against BOD₅, COD, color, and ammoniacal nitrogen. The RO membrane, in particular, achieves at least 95% removal for all the general parameters. The figure also shows that all of the membranes are able to achieve almost complete elimination of suspended solids. The excellent results are due to the significantly smaller membrane pore dimension compared to the particle size of the suspended solids (>2 μ m).

When it comes to the metal-based parameters (Fig. 5(b)), one can observe the high efficiency of the NF and RO membranes in removing metallic elements compared to the loose MF/UF membranes. The presence of many metal ions in the leachate is emanated by the solid wastes disposed in landfills that are caused by the overwhelming consumption of high-tech products in the modern era. The high separation efficiency of the NF and RO membranes is confirmed through the low concentration of the respective elements detected in the permeate samples. The efficiencies of NF/RO membranes in rejecting metallic elements are in good agreement with their high rejection rates against NaCl as shown in Fig. 1.

Further analysis also indicates that the NF and RO membranes are able to reduce the values of parameters such as cyanide, fluoride, formaldehyde, phenol, and sulfide by achieving low concentrations of <0.01 mg/L, 0.2-0.3 mg/L, 0.9 mg/L, <0.001 mg/L and $\le 0.1 \text{ mg/L}$, respectively in the permeates. In comparison to the RO membrane, the NF membrane is found to demonstrate significantly lower capability in removing boron (atomic number: 5), iron (atomic number: 26), and selenium (atomic number: 34). These three elements are also found to present at relatively high concentration in the local leachate. Typically, boron, which exists in the form of boric acid at pH 7.5-8, can only be rejected by commercial seawater RO membranes at 83-92% [25]. The boron rejection rate by the RO membrane is even lower compared to NaCl rejection. Thus, the presence of a higher concentration of boron (compared to other metallic elements) in the permeate produced by the NF and RO membrane is reasonable. Other metallic elements, i.e., mercury, cadmium, chromium hexavalent, arsenic, lead, copper, and silver are not shown in this figure as their concentrations are very low (not detected by the analytical methods employed in Table 3) in the landfill leachate.

The performance of the membranes in removing fluoride, formaldehyde, sulfide, and phenol from the leachate is also assessed and the findings are presented in Fig. 5(c). In general, the NF and RO membranes demonstrate consistent performance in removing these compounds by producing the permeates with very low concentrations. The concentration of these four compounds detected in the permeate of NF and RO membranes is more or less the same. However, their performance is obviously better than those of loose MF/UF membranes, particularly in removing fluoride and formaldehyde. The concentration of cyanide (CN-) is not presented in the figure due to its extremely low concentration in the landfill leachate (<0.01 mg/L).

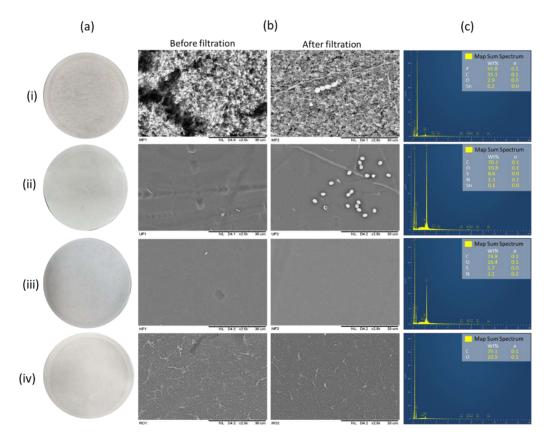


Fig. 4. (a) Surface photograph of membranes (diameter: 5 cm) after being used for leachate treatment, (b) SEM surface images of membrane before and after leachate treatment, and (c) EDX analysis on the surface chemistry of used membranes. (i) MF, (ii) UF, (iii) NF, and (iv) RO membranes

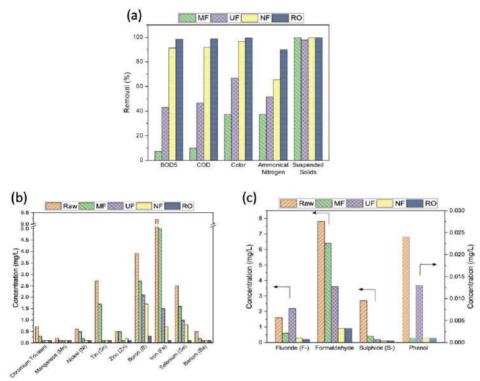


Fig. 5. In-depth analysis of the quality of leachate treated by different membranes, (a) general parameters, (b) metal-based parameters, and (c) other parameters (Note: pH of raw leachate: 8.4; pH of treated leachate: 8.6 (MF), 8.5 (UF), 8.6 (NF), and 9.1 (RO))

Table 4.Comparison of the best-performing membrane studied in this work with other membranes reported in the literature for leachate treatment

	In this work		Kosutic et al. [27]		Chaudhari & Murthy [28]		Elfilali et al. [29]	
Parameter	^a Feed (mg/L)	bRO permeate (mg/L)	^a Feed (mg/L)	bNF270 permeate (mg/L)	^a Feed (mg/L)	b,cNF-300 permeate (mg/L)	^a Feed (mg/L)	dMBR/UF permeate (mg/L)
pН	8.42	8.27	8.05	7.87	6.8	_	8.14	7–8
BOD_5	2100	30 (98.57%)	_	-	18,603	2952 (84.13%)	895-1250	116.5–162.5 (87%)
COD	3000	40 (98.68%)	1720	67 (94.6%)	56,521	1678 (97.03%)	4985-7433	1196–1784 (76%)
Color	213	1 (99.53%)	Dark brown	No color	150	24 (84%)	Dark brown	Brown
N	3395	340 (89.99%)	1147.6	664.5 (37.1%)	196	59 (69.90%)	725-1025	292.2-413 (59.7%)
Sn	2.7	0.1	0.0452	0.0002	-	-	-	-
Zn	0.5	0.1	0.106	0.0089	0.188	0.0038	-	-
В	3.9	0.3	7.36	7.17	-	-	-	-
Fe	9.2	< 0.1	6.41	0.0355	-	-	-	-
Se	2.5	< 0.1	0.0207	0.0165	-	-	-	-
F-	2.7	0.2	19.59	0.93	0.4	0.088	-	-

^a All parameters are in the unit of mg/L, except pH and color (ADMI)

^d The rejection (%) shown in the bracket is the average of the result.

Table 4 presents a comparison between the most effective RO membrane studied in our work and other membranes documented in the literature for leachate treatment. Clearly, when the primary consideration is the quality of the permeate, the RO membrane stands out as the superior choice. It exhibits a rejection rate of at least 98.57% against parameters like BOD₅, COD, and color. This rejection rate is notably higher when contrasted with alternative membrane types such as NF, which range from 84% to 97.03%, and UF, which falls within the range of 76% to 87% when employed for leachate treatment. Furthermore, the RO membrane displays considerable potential for nitrogen removal and generates permeate with only trace amounts of metallic elements.

By comparing the quality of the treated leachate with the local regulation (see Table 1), it is found that NF and RO membranes are the membranes that can meet most of the discharge standards. Both membranes achieve better results for all metallic removals in comparison to the discharge standard of leachate, except for boron parameters (NF membrane) which records 1.7 mg/L – slightly higher than the standard (1.0 mg/L). Furthermore, both NF and RO membranes also achieve superior results for suspended solids, COD, and color compared to the discharge standard. Nevertheless, it must be pointed out that both membranes fail to achieve good results for BOD $_5$ (Max: 20 mg/L) and ammoniacal nitrogen (Max: 5 mg/L). Their values for BOD $_5$

and ammoniacal nitrogen are relatively high, recording at $30{\text -}185~\text{mg/L}$ and $340{\text -}1169~\text{mg/L}$, respectively. The main factor contributing to the inefficiency of BOD_5 and ammoniacal nitrogen removal is the use of freshly collected leachate for the filtration experiment. This leachate sample did not undergo any biological treatment to reduce the amount of biodegradable organic matter. Typically, leachate is required to undergo a proper aerobic/anaerobic treatment in order to sufficiently reduce the level of BOD_5 and ammoniacal nitrogen prior to the next treatment process. Besides the biological pretreatment, researchers also reported that coagulation-flocculation (C/F) using ferric chloride (FeCl₃) and alum sulfate (Al₂(SO₄)₃) prior to the membrane could offer a good solution to mitigate membrane fouling and minimize severe flux deterioration during leachate treatment [20].

4. Conclusions

Currently, there is no landfill in Malaysia employing membrane technology in treating the leachate. Our current work is the first such study that investigated the efficiency of different classes of polymeric membranes ranging from MF to RO for landfill leachate treatment and carried out a comprehensive assessment of the quality of treated water according to the

^b The number in the bracket indicates the removal efficiency (%).

^cThe data presented were obtained from the filtration experiment carried out at 20 bar.

local environmental quality regulations. Our findings revealed that although the UF and MF membranes displayed remarkably greater water permeability than those of RO and NF membranes during the landfill leachate treatment, the quality of the treated water in terms of BOD5, COD, color and ammoniacal nitrogen is far below the quality of water treated by the NF and RO membranes. Furthermore, the dense NF and RO membranes could produce the permeates with low concentrations for all metallic elements present in the leachate, except for boron, iron, and selenium which existed at relatively high concentrations in the leachate. By comparing the NF and RO membranes, our experimental results showed that the RO membrane is superior in producing higher-quality treated leachate. Besides demonstrating higher efficiencies in reducing the levels of general and metallic-based parameters, the RO membrane also displayed excellent results in removing fluoride and sulfide from the leachate. Nonetheless, it must be pointed out that since this study only evaluated the performance of individual membranes without considering the hybrid process and the raw leachate sample was freshly collected without having any pre-treatment (e.g., biological process), it is rather difficult to meet all the parameters of discharge standards, especially BOD₅ and ammoniacal nitrogen which are greatly dependent on the biological pre-treatment process. Thus, it is highly recommended to integrate the NF/RO membrane with suitable pre-treatment processes (biological and physical treatments) to achieve a higher quality of final discharged leachate while improving the water permeability of the membranes.

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Data availability

Data are available upon request.

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CRedit authorship contribution statement

- Y. K. Tan: Conceptualization; Funding acquisition; Resources; Writing review.
- W.J. Lau: Conceptualization; Funding acquisition; Data curation; Writing original draft; Formal analysis; Supervision
- N. S. M.Nawi: Investigation; Methodology; Formal analysis; Visualization
- R. A. Roslan: Investigation; Methodology; Formal analysis
- P. S. Ng: Conceptualization; Resources; Writting review & editing

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

- International Trade Administration, Malaysia Waste Management Solutions, (2022). https://www.trade.gov/market-intelligence/malaysiawaste-management-solutions (accessed April 28, 2023).
- [2] M.A. Hannan, Md. Abdulla Al Mamun, A. Hussain, H. Basri, R.A. Begum, A review on technologies and their usage in solid waste monitoring and management systems: Issues and challenges, Waste Manag. 43 (2015) 509–523. https://doi.org/10.1016/j.wasman.2015.05.033.
- [3] S.T. Tan, C.T. Lee, H. Hashim, W.S. Ho, J.S. Lim, Optimal process network for municipal solid waste management in Iskandar Malaysia, J. Clean. Prod. 71 (2014) 48–58. https://doi.org/10.1016/j.jclepro.2013.12.005.
- [4] T. Abunama, F. Othman, T.I.T. Nilam, Comparison of landfill leachate generation and pollution potentials in humid and semi-arid climates, Int. J.

- Environ. Waste Manag. 27 (2021) 79–92. https://doi.org/10.1504/ijewm.2021.111906.
- [5] R.B. Brennan, M.G. Healy, L. Morrison, S. Hynes, D. Norton, E. Clifford, Management of landfill leachate: The legacy of European Union Directives, SISanitary Landfilling. 55 (2016) 355–363. https://doi.org/10.1016/j.wasman.2015.10.010.
- [6] L. Alibardi, R. Cossu, 6.1 Leachate Generation Modeling, in: R. Cossu, R. Stegmann (Eds.), Solid Waste Landfilling, Elsevier, 2018: pp. 229– 245. https://doi.org/10.1016/B978-0-12-407721-8.00013-9.
- [7] M.A. Kamaruddin, M.S. Yusoff, L.M. Rui, A.M. Isa, M.H. Zawawi, R. Alrozi, An overview of municipal solid waste management and landfill leachate treatment: Malaysia and Asian perspectives, Environ. Sci. Pollut. Res. 24 (2017) 26988–27020. https://doi.org/10.1007/s11356-017-0303-9.
- [8] S. Renou, J.G. Givaudan, S. Poulain, F. Dirassouyan, P. Moulin, Landfill leachate treatment: Review and opportunity, J. Hazard. Mater. 150 (2008) 468–493. https://doi.org/10.1016/j.jhazmat.2007.09.077.
- [9] Q.-Q. Zhang, B.-H. Tian, X. Zhang, A. Ghulam, C.-R. Fang, R. He, Investigation on characteristics of leachate and concentrated leachate in three landfill leachate treatment plants, Waste Manag. 33 (2013) 2277– 2286. https://doi.org/10.1016/j.wasman.2013.07.021.
- [10] H. Li, H. Liu, Treatment and recovery methods for leachate concentrate from landfill and incineration: A state-of-the-art review, J. Clean. Prod. 329 (2021) 129720. https://doi.org/10.1016/j.jclepro.2021.129720.
- [11] F.N. Ahmed, C.Q. Lan, Treatment of landfill leachate using membrane bioreactors: A review, Desalination. 287 (2012) 41–54. https://doi.org/10.1016/j.desal.2011.12.012.
- [12] R. Keyikoglu, O. Karatas, H. Rezania, M. Kobya, V. Vatanpour, A. Khataee, A review on treatment of membrane concentrates generated from landfill leachate treatment processes, Sep. Purif. Technol. 259 (2021) 118182. https://doi.org/10.1016/j.seppur.2020.118182.
- [13] H. Luo, Y. Zeng, Y. Cheng, D. He, X. Pan, Recent advances in municipal landfill leachate: A review focusing on its characteristics, treatment, and toxicity assessment, Sci. Total Environ. 703 (2020) 135468. https://doi.org/10.1016/j.scitotenv.2019.135468.
- [14] J. Zhang, K. Xiao, X. Huang, Full-scale MBR applications for leachate treatment in China: Practical, technical, and economic features, J. Hazard. Mater. 389 (2020) 122138. https://doi.org/10.1016/j.jhazmat.2020.122138.
- [15] K. Cirik, S. Gocer, Performance of anaerobic membrane bioreactor treating landfill leachate, J. Environ. Health Sci. Eng. 18 (2020) 383–393. https://doi.org/10.1007/s40201-019-00376-9.
- [16] E. Coppini, L. Palli, D. Fibbi, R. Gori, Long-term performance of a full-scale membrane plant for landfill leachate pretreatment: a case study, Membranes. 8 (2018). https://doi.org/10.3390/membranes8030052.
- [17] M. Altinbas, H. Ozturk, E. İren, Full-scale sanitary landfill leachate treatment by MBR: flat sheet vs. hollow fiber membrane, J. Membr. Sci. Res. 7 (2021) 118–124. https://doi.org/10.22079/jmsr.2020.123563.1358.
- [18] S. Chen, J. Liu, Landfill leachate treatment by MBR: Performance and molecular weight distribution of organic contaminant, Chin. Sci. Bull. 51 (2006) 2831–2838. https://doi.org/10.1007/s11434-006-2177-y.
- [19] M. El-Fadel, J. Hashisho, A comparative examination of MBR and SBR performance for the treatment of high-strength landfill leachate, J. Air Waste Manag. Assoc. 64 (2014) 1073–1084. https://doi.org/10.1080/10962247.2014.907840.
- [20] N. Remmas, P. Melidis, G. Paschos, E. Statiris, S. Ntougias, Protozoan indicators and extracellular polymeric substances alterations in an intermittently aerated membrane bioreactor treating mature landfill leachate, Environ. Technol. 38 (2017) 53–64. https://doi.org/10.1080/09593330.2016.1190792.
- [21] Department of Environment (DOE) Malaysia, Environmental quality (pollution control from solid waste transfer stations and landfills) regulations 2009 -P.U. (A) 433/2009, (2009). https://www.doe.gov.my/en/environmental-quality-pollution-controlfrom-solid-waste-transfer-stations-and-landfills-regulations-2009-p-u-a-433-2009 (accessed April 28, 2023).
- [22] D. Roy, A. Azaïs, S. Benkaraache, P. Drogui, R.D. Tyagi, Composting leachate: characterization, treatment, and future perspectives, Rev. Environ. Sci. Biotechnol. 17 (2018) 323–349. https://doi.org/10.1007/s11157-018-9462-5.
- [23] W.J. Lau, A.F. Ismail, S. Firdaus, Car wash industry in Malaysia: Treatment of car wash effluent using ultrafiltration and nanofiltration membranes, Sep. Purif. Technol. 104 (2013) 26–31. https://doi.org/10.1016/j.seppur.2012.11.012.
- [24] C.S. Ong, W.J. Lau, A.F. Ismail, Treatment of dyeing solution by NF membrane for decolorization and salt reduction, Desalination Water Treat. 50 (2012) 245–253.

- [25] Z.C. Ng, C.Y. Chong, W.J. Lau, M. Karaman, A.F. Ismail, Boron removal and antifouling properties of thin-film nanocomposite membrane incorporating PECVD-modified titanate nanotubes, J. Chem. Technol. Biotechnol. 94 (2019) 2772–2782. https://doi.org/10.1002/jctb.6044.
- [26] N.K. Saha, S.V. Joshi, Performance evaluation of thin film composite polyamide nanofiltration membrane with variation in monomer type, J. Membr. Sci. 342 (2009) 60–69. https://doi.org/10.1016/J.MEMSCI.2009.06.025.
- [27] K. Košutić, D. Dolar, T. Strmecky, Treatment of landfill leachate by membrane processes of nanofiltration and reverse osmosis, Desalination Water Treat. 55 (2015) 2680–2689. https://doi.org/10.1080/19443994.2014.939863.
- [28] L.B. Chaudhari, Z.V.P. Murthy, Treatment of landfill leachates by nanofiltration, J. Environ. Manage. 91 (2010) 1209–1217. https://doi.org/10.1016/j.jenvman.2010.01.007.
- [29] N. Elfilali, N. Essafi, M. Zait, M. Tahaikt, F. Elazhar, A. Elmidaoui, M. Taky, Effectiveness of membrane bioreactor/reverse osmosis hybrid process for advanced purification of landfill leachate, Desalination Water Treat. 240 (2021) 24–32. https://doi.org/10.5004/dwt.2021.27560.