Journal of Emerging Technologies and Industrial Applications
Vol. 4 No. 2 (2025) pp. 1-6
e-ISSN: 2948-507X
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Journal homepage: https://jetia.mbot.org.my/index.php/jetia/index

Improvement of biological treatment of leachate using membrane bioreactor (MBR)

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Abstract: A laboratory scale of Membrane bioreactor (MBR) was used to treat leachate that collected from Pekan Nenas Sanitary Landfill. Before MBR treatment, the suitable pore size of membrane was identified to use in MBR treatment by membrane filtration. As the treatment started, there is a cake layer formation. The layer was analysed using SEM. The findings of this research showed that the 600 Dalton pore size of membrane can filter clearer leachate colour. The SEM analyses showed the small particles in the top layer indicate that small flocs had a strong tendency to deposit on membrane surface.

Received 15 June 2025; Accepted 21 September 2025; Available online 28 December 2025

Keywords: Pore size, membrane bioreactor (MBR), colour, cake layer, filtration

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1. Introduction

The major permanent problems caused by landfills correspond to the generation of leachate, which can lead to significant environmental challenges [1]. Climate, the age of the landfill, solid waste composition, and the hydrology of the landfill are key factors influencing the composition of landfill leachate. Landfill leachate is defined as any liquid that percolates from solid waste and extracts dissolved, suspended, or microbial contaminants from it [2],[3]. Organic compounds, nutrients, heavy metals, suspended solids, and nitrogen are the main pollutants that need to be removed during leachate treatment. High nitrogen concentrations and leachate toxicity limit the effectiveness of typical activated sludge processes in continuous flow reactors [4],[5] However, the most persistent issue in leachate treatment is the removal of humic substances (HS), which contribute to the colour of leachate.

HS, including humic acid (HA) and fulvic acid (FA), are byproducts of the natural decay of plant and animal materials [6]. These compounds play significant roles in pollutant chemistry and biogeochemistry but are challenging to remove due to their complex molecular structures [7]. HS are major contributors to flux decline during filtration [8] and form hazardous metal complexes and carcinogenic substances during chlorination. According to the World Health Organization (WHO), the permissible HA concentration in potable water is about 100 ppb [3],[9]. Effective removal of HS, particularly their contribution to colour, is a critical focus in leachate treatment.

Various methods, such as electrocoagulation, reverse osmosis (RO), and membrane filtration, have been explored for colour removal. While RO is highly effective, it is costly and impractical for large-scale systems [10]. Membrane bioreactors (MBRs), which replace sedimentation tanks with filtration systems, offer a cost-effective and space-saving alternative. The pore size of the membrane is crucial for pollutant removal, particularly for HS, which can contribute to odour, taste, and acidity problems and are precursors for trihalomethane formation [11],[12]. However, the unpredictable molecular weight of HS and membrane fouling caused by cake layer formation remain major challenges [13].

Membrane fouling, resulting from the accumulation of pollutants on the membrane surface, reduces filtration performance. Cake layer formation, in particular, increases filtration resistance and can significantly impact the transmembrane pressure and flux rate [14]. Understanding the influence of cake formation on membrane performance, as well as identifying the optimal pore size for effective filtration, is essential for improving leachate treatment. In this study, a comparison of membrane filtration and MBR performance is conducted to evaluate the efficiency of both systems in removing pollutants such as COD, BOD, NH3-N, and colour, with a focus on the role of pore size and cake layer formation. Thus, the objectives of this study are (a) to determine the optimal membrane pore size for leachate filtration; (b) to compare the efficiency of membrane filtration with Membrane Bioreactor (MBR) system and (c) to analyse the impact of cake layer formation on membrane performance.

2. Materials and Methods

In this research, various chemicals and equipment were used. Different methods were carried out to analyze each parameter contained in the leachate. The preparation for this experiment was simplified into three phases. Phase one involved the design of the system. Phase two focused on the procurement phase, where the system was built. Lastly, phase three dealt with commercialization and testing.

2.1 Membrane Filtration

For this filtration, 3 different pore size of membrane was filter using Sterlitech HP4750 Stirred Cell as shown in Fig. 1. The leachate that collected for Pekan Nenas sanitary landfill was used. The pore size of membrane was 600 Da, 20 kilo Da and 6000 kilo Da respectively as shown in Table 1. The features and technical specification of HP4750 is shown in Table 2.

Table 1 - Specification of membrane

Pore size	Type of material	Type of filtration
600 Da	Thin film composite	Nano-filtration (NF)
20 kDa	Polysulfone	Ultrafiltration (UF)
6000 kDa	Polyvinylidene fluoride	Microfiltration (MF)

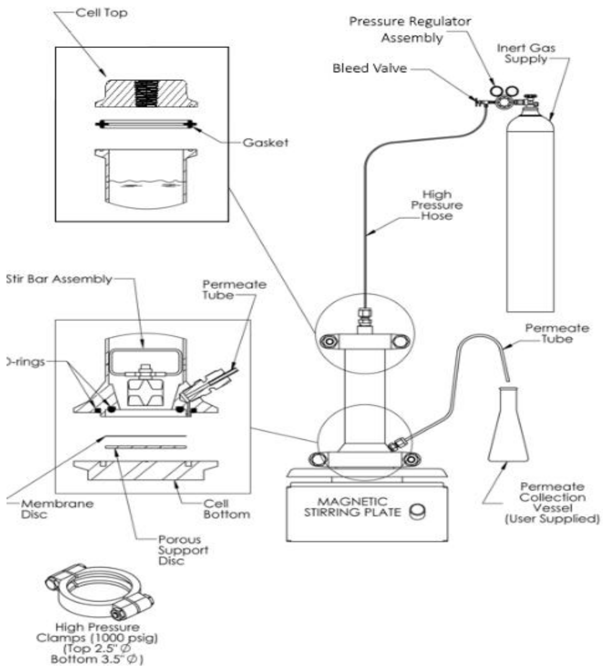

Fig. 1 Components of HP4750

Table 2 - Features and technical specification of HP4750

Parameter	Description
Membrane size	47-49mm diameter
Active membrane area	14.6 cm^2
Processing volume	300 Ml
Hold-up volume	1 Ml
Maximum pressure	69 bar
pH range	Membrane dependent

2.2 MBR System

The system constructed in pilot scale for this research. The pore size of membrane that used in MBR was 6000 kilo Da. The specification for the membrane was shown in Table 2. The system was construct without the sedimentation tank. The sedimentation tank was replaced by the membrane tank. The effluent was tested. COD, BOD, color, ammoniacal nitrogen and pH was test with different method. COD was test by HACH Method 8000. The BOD was measured by conducting BOD_5 test. The color changed was measured using Platinum-Cobalt Standard Method. The ammoniacal nitrogen was tested by HACH Method 10031. Lastly for pH, pH meter was used. The diagram of designed treatment system and specification of MBR system is shown in Fig. 2 and Table 3 respectively.

Table 2 - Specification of membrane used for MBR

Material	Polyvinylidene fluoride (PVDF)
Type	Hollow fiber
Pore Structure	Slit pore
Nominal pore size	6000kDa
Outer diameter / inner diameter	1.2mm / 0.7mm
Operating flux	25LMH

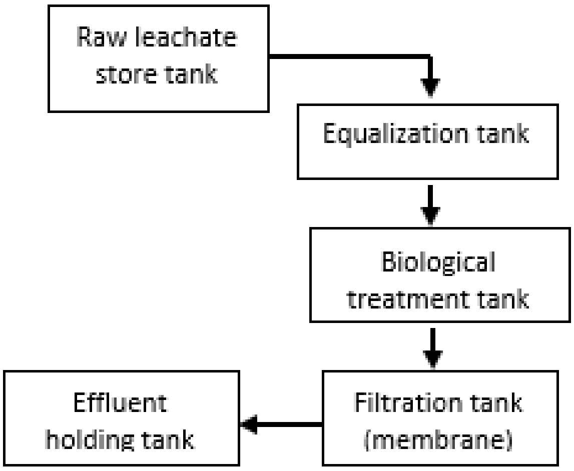

Fig. 2 - Diagram of designed treatment system

Table 3 - Specification of MBR system

Volume of tank	52 L
Type of container	Plastic container (3 Units)
Diameter of pipe	½ inch
Flowrate	6.5 L/hr

2.3 Cake Layer Analysis

After the MBR treatment run for approximately 1 month 18 days, slimy layer formed on the surface of membrane. The slimy layer was analyzed using Scanning Electron Microscope (SEM). The equipment that used was SEM model JEOL JSM-6390LV. This SEM produced result of topography of membrane surface by scanning the surface with a focused beam of electrons. The membrane layer (1 cm × 1 cm) was cut into 2 pieces with cake layer on its surface. In order to prevent the structure and thickness of cake layer from change, the cake layer was coated with layer of platinum approximately 2 nm thick using BAL-TEC SCD 050 Sputter Coater. The analysis was carried out in vacuum conditions (1×10^{-8} Pa) at an accelerating voltage of 15 kV.

3. Results and Discussions

3.1 Membrane Filtration

The leachate was collected from the Pekan Nenas sanitary landfill during normal days. The raw leachate was analyzed before starting the experiment. The concentrations of BOD, COD, ammoniacal nitrogen, pH, and color in the leachate were tested. Table 4 represents the composition of the leachate collected.

Table 4 Results of the raw leachate analysis

Parameters	Raw leachate	Standards**
COD (mg/L)	3900	400
BOD_5 (mg/L)	260.7	20
NH_3-N (mg/L)	3060	5
pH	8.46	6.0-9.0
Color (PCU)	15000	100 (ADMI)*

From the results, the Pekan Nenas Sanitary Landfill is categorized as an old landfill. An old landfill is defined as a landfill that has been operating for more than 10 years. This statement is verified by the results obtained from the leachate analysis. The analysis shows that the pH value is 8.46 and the ammoniacal nitrogen concentration is 3060 mg/L. A typical old landfill leachate is usually more alkaline and contains ammoniacal nitrogen exceeding 400 mg/L. This is further confirmed as the Pekan Nenas Sanitary Landfill had been in operation for 8 years since 2006. The average waste received by the landfill was 160 tons/day. This landfill ceased operations in 2013 and now transfers waste to the Seelong sanitary landfill.

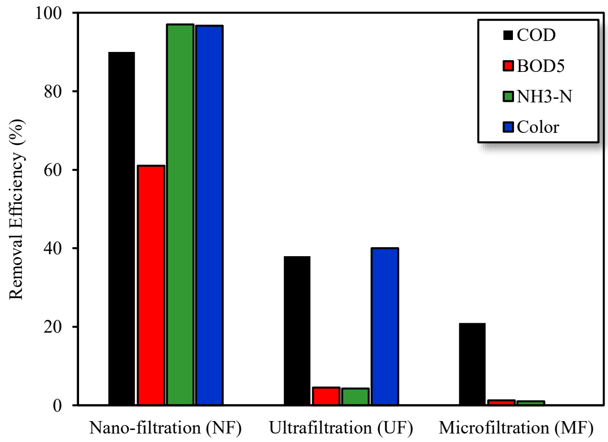

Fig. 3 – Removal efficiency of raw leachate parameters by using different type of membrane

Fig. 3 shows removal efficiency of raw leachate parameters by using different type of membrane. According to the results, a 600 Da nanopore size membrane can reduce the color of leachate by 96.7%. This proves that nanopore size membranes filter leachate color better than ultra-pore size and micropore size membranes. However, using a 600 Da membrane in a leachate treatment system requires a pressure of 15 bar. Even though this pressure was supplied in the experiment, it took 26.45 minutes to filter just 7 mL of leachate. In a real plant, a larger surface area of the nanopore membrane would be required. A larger surface area allows the membrane to filter leachate in larger

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quantities more quickly. On the other hand, using a pressure of 15 bar frequently damages the membrane, increasing costs. To reduce costs and save space in the treatment plant, a membrane bioreactor (MBR) was chosen.

3.2 Evaluation of Membrane Flux

Fig. 4 show flux of each membrane. It can be concluded that the flux rate of each membrane decreases over time. The standard operating flux for nanofiltration is 8–10 L/m². hr.bar. The average flux obtained through the experiment was 8.544 L/m². hr.bar. When the calculated flux value is compared to the standard operating flux, it falls within the range, indicating that the flux value obtained during filtration is reliable. The operating standard flux reflects the membrane's behavior in filtering the solution.

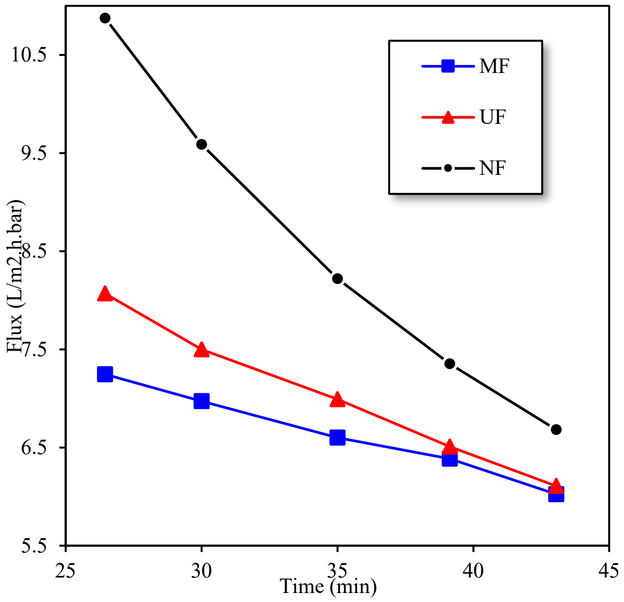

Fig. 4 - Flux of each membrane

Table 5 Comparison of flux with operating flux

Membrane pore size	Standard operating flux (L/m^2.hr.bar)	Flux obtains ((L/m^2.hr.bar)
600 Da	8-10	8.5
20 kDa	150	7.0
6000 kDa	300	6.6

For ultrafiltration, the standard operating flux was 150 L/m².hr.bar. The average flux obtained through the experiment was 7.035 L/m².hr.bar. According to Mulder (1996), pure water permeability for ultrafiltration membranes typically ranges from 10–50 L/m².h.bar for operating pressures of 1–5 bars. There is a significant difference between the operating flux and the flux obtained during the experiment. This discrepancy might be due to the type of filtration conducted. Typically, two major competing filtration technologies are used in industrial processes: dead-end filtration and cross-flow filtration. In dead-end filtration, solids collect on the surface of the filter media, forming a stable filter cake that grows in thickness and increases flow resistance. The filter media is usually a consumable component disposed of along with the filtered solids. In contrast, in cross-flow filtration, the concentrate feed water flows across the membrane filter media's surface with minimal solids buildup and constant low flow resistance. The membrane is not a consumable component. The flux decline of relatively high-flux membranes (e.g., ultrafiltration) can be influenced by natural organic matter (NOM) aromaticity and membrane hydrophobicity.

Since fouling results in flux reduction and subsequently increases operational costs, it is essential to develop membranes with antifouling properties. To promote this characteristic and reduce fouling, modification of the membrane surface is necessary. The primary aim of surface modification is to increase hydrophilicity, alter wettability, or fabricate an antimicrobial surface. Hydrophilicity refers to the tendency of a molecule to interact favorably with water and other polar substances rather than oil or hydrophobic solvents. Hydrophilic molecules are typically charge-polarized and capable of hydrogen bonding. One study demonstrated the modification of a reverse osmosis (RO) membrane surface with silver nanoparticles on both the membrane and the spacer. The results revealed that the coated membranes and spacers exhibited better antimicrobial activity, resulting in a more moderate flux decline. Research has shown that membrane hydrophilicity and hydrophobicity significantly affect flux decline during membrane filtration [20]. Recent studies have also reported that sludge adhesion, which leads to the formation of a cake layer, primarily results from interfacial interactions between sludge foulants and the membrane surface. While hydrodynamic forces drive the foulants closer to the membrane surface, it is the short-range interfacial interaction forces that ultimately bind the foulants to the membrane.

3.3 MBR Treatment

Fig. 5 shows colour removal efficiency by using MBR at different batches. Since supplying a pressure of 15 bar in a real treatment plant is impractical, a membrane with a pore size of 6000 kDa was used with a biological system at a supplied pressure of 1 bar. This system is referred to as a membrane biological system, where the sedimentation unit is replaced by membrane filtration. The sedimentation unit occupies a significant amount of space in treatment plants. Alternatively, using membranes eliminates the need for a sedimentation tank, saving space for building the treatment plant. Seven batches of treatment were carried out using five sets of raw leachates. The effluent filtered through the membrane utilized gravitational force. The membrane was arranged as a side-stream membrane to the

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biological tank. The color changes in each batch of filtration were observed. The treatment continued until a cake layer formed on the membrane surface, causing membrane fouling.

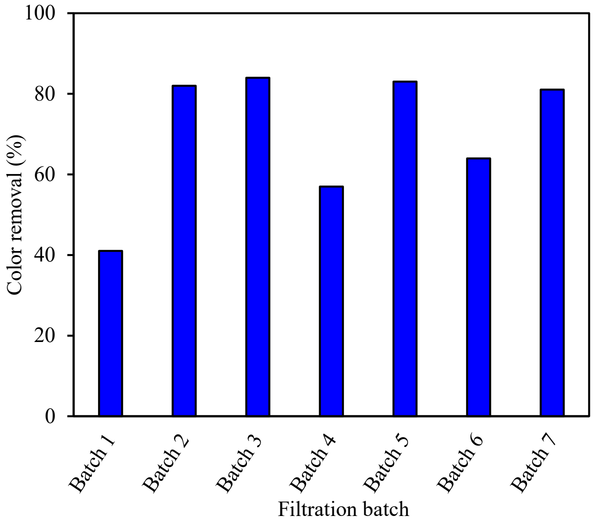

Fig. 5 - Result of seven batch of MBR treatment

Overall, the optimum color reduction was achieved during the treatment of Batch 5, which used a flow rate of 119.232 L/day. Observations with the naked eye also confirmed that the effluent from the membrane tank was lighter in color. Therefore, it can be concluded that the optimum flow rate is 119.232 L/day. Membrane fouling began during the treatment of Batch 6. This is evident as the color reduction efficiency decreased from 85.05% in Batch 5 to 67.68% in Batch 6. Chemical cleaning is necessary to restore the membrane's efficiency for continued treatment.

3.4 Cake Layer Analysis

The SEM images of the cake layer provide valuable insights into its structure and its impact on membrane performance. The top cake layer, as shown in the Fig. 6, is characterized by a dense and compact structure with larger particle sizes ranging from 47.49 µm to 60.00 µm. This dense layer is formed by the deposition of larger foulants and suspended solids during filtration, creating a primary barrier to flow. The compactness of this layer increases filtration resistance and leads to a decline in flux over time, necessitating higher transmembrane pressure (TMP) to maintain filtration performance. Previous experiments demonstrated that the operational pressure reached 15 bar, illustrating the substantial resistance caused by this dense top layer. The rapid formation of this layer emphasizes the need for frequent cleaning cycles to sustain membrane performance.

In contrast, the bottom cake layer, shown in the Fig. 7, is more porous and less compact, with particle sizes ranging from 33.14 µm to 52.00 µm. This layer reflects early-stage fouling, where smaller particles settle and bind with the membrane surface. Although less resistant than the top layer, the bottom layer contributes to irreversible fouling due to short-range interfacial interactions between foulants and the membrane surface. Over time, the bottom layer integrates with the top layer, further exacerbating the decline in flux and filtration efficiency.

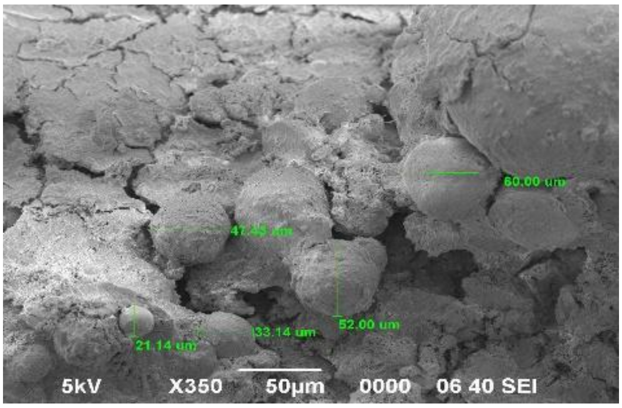

Fig. 6 - SEM image of top cake layer

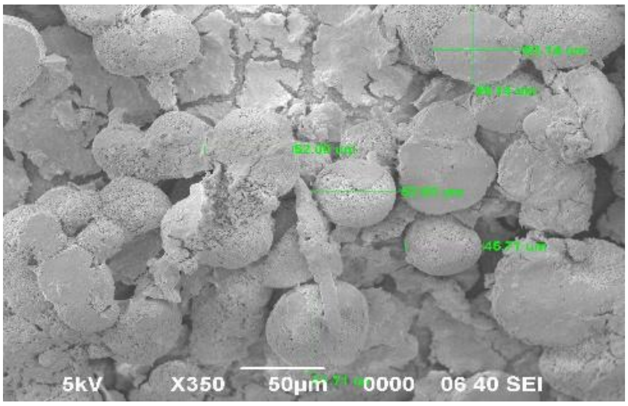

Fig. 7 - SEM image of bottom cake layer

The numerical data support these observations. The average flux for nanofiltration was 8.544 L/m².hr.bar, within the standard operating range of 8–10 L/m².hr.bar, confirming reliable performance under controlled conditions. However, for ultrafiltration, the flux dropped to 7.035 L/m².hr.bar, significantly lower than the standard range of 10–50 L/m².hr.bar, likely due to the impact of the cake layer. Additionally, the color removal efficiency declined as fouling progressed. Batch 5 achieved an optimal color reduction of 85.05% at a flow rate of 119.232 L/day, but Batch 6 experienced reduced efficiency at 67.68%, attributed to the increased thickness and compaction of the cake layer.

The stratification of the cake layer demonstrates the significant impact of fouling on membrane performance. The dense top layer contributes primarily to reversible fouling, which can be mitigated through chemical cleaning, while the porous bottom layer leads to irreversible fouling that affects the long-term usability of the membrane. To address these challenges, operational parameters such as flow rate and pressure must be optimized to delay cake layer formation. Enhanced cleaning protocols are also necessary to manage both reversible and irreversible fouling. Furthermore, surface modification of membranes to reduce interactions with foulants can mitigate adhesion and prolong membrane lifespan. In conclusion, the cake layer significantly influences filtration resistance, flux decline, and pollutant removal efficiency, emphasizing the need for integrated strategies to manage fouling and enhance membrane performance.

4. Conclusion

From the analysis results, the leachate from Pekan Nenas Sanitary Landfill resembles the composition of leachate generated from old landfill. In the membrane filtration experiment, the highest removal of COD, BOD5, colour and ammoniacal nitrogen was using nano pore size membrane. It can be reduced to about 92.3% of COD, 62.2% of BOD5, 96.7% of colour and 96.3% of ammoniacal nitrogen. For MBR system, batch 5 showed optimum performance with reduction 99.32% of COD, 91.62% of BOD5, 69.33% of ammoniacal nitrogen and 85.05% of colour. Even though MBR system effluent colour did not comply with standard discharge of American Dye Manufacturer's Institute, this system is more convenient and practical to implement in real leachate treatment plant compare to membrane filtration only.

Acknowledgement

This work was supported by the Universiti Teknologi Malaysia (UTM) Fundamental Research Grant (Grant No. Q.J130000.3846.22H85).

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