Citrus Fruit Waste Leachate Treatment By Using Newly Developed Flat Sheet Membrane

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Abstract— The increasing of the population has enhanced the generation of waste in Malaysia, a country of which depends on landfill as a method to dispose of solid waste in which the leachate generation will play a major impact to the environment. Leachate can potentially contaminate the nearby surface water, groundwater and soil. In Malaysia, food waste is a major waste composition which can lead to the generation of leachate due to their high moisture content. Citrus fruit wastes classified as one of food waste which become a concern because of its potential to create environmental pollution. Leachate from citrus fruit is acidic, high BOD and COD, high turbidity, total suspended solids and high nutrient such as nitrogen and phosphorus. This characteristic may interfere the effectiveness of wastewater treatment. The method that can be implemented as citrus leachate treatment is membrane process. Membrane process had been proven their effectiveness and had been used in many applications. The previous membrane composition is used, but the suitability of citrus waste is needed to investigate. The MWCO of newly developed flat sheet membrane is determined to study the performance of the membrane in citrus fruit waste leachate treatment.

Index Terms- Citrus Fruit Waste, Leachate, Membrane Process

I. INTRODUCTION

Malaysia is a country which highly depends on landfill as a method to dispose of solid waste. However, due to the generation of leachate which can potentially contaminate nearby surface and groundwater if left untreated, landfills require close environmental monitoring [1]. Leachate can be defined as the aqueous effluent as the results from the percolation of rainwater through waste, a biochemical process in waste's cell and water content from the waste themselves [2]. During the percolation process through the waste, it will also carry along the organic, inorganic, heavy matter, pathogen and other polluted matter. Therefore, leachate has a great environmental concern due to its potential to create pollution. Although there are many different characterisations of solid waste in Malaysia, the primary constituent is food or organic waste which contributes up 37.43% [3].

Landfills are known as the primary treatment of food waste in all developing countries which estimated to be around 90% of total food waste disposal [4]. However, due to its biodegradability, landfill practice is not considered as a feasible method for food waste treatment and existing of food waste in landfills can result in disease vectors. One of the types of food waste that can contribute to the production of leachate is fruit waste. Due to its nutritious contents such as vitamin and various applications, fruit is consumed widely in the world and hence produce a significant amount of fruit waste mainly the fruit peel which generated from fruit processing industries [5]. Citrus fruit is one of the types of fruit waste that can easily be found in Malaysia. The waste generated from the production of citrus fruits can become a great environmental if they are not correctly being managed [6]. Besides that, the effluent of citrus wastewater has a very high organic load which BOD is 20-1400 mg/l and COD is 100-2000 mg/l [7]. Citrus effluent is high variability of the low value of pH which is usually acidic [7]. Therefore, it is essential to treat the leachate from citrus fruit waste to avoid the environmental problem and their effects on the wastewater treatment.

Numerous approaches had been applied to the leachate treatment. For many years, the conventional biological treatments and classical physicochemical method are considered as the most appropriate technologies for management and manipulation of high strength effluent such as landfill leachate [2]. However, these methods are not sufficient anymore in reducing the negative impact of the landfill leachate on the environment as the continuous hardening of the discharge standard in most of the countries and the ageing of landfill sites with more stabilised leachate. Treatment based on membrane technologies has emerged as the alternative treatment to improve the current methods and to comply with water quality regulation in most countries. Membrane processes are considered as the new treatment of leachate which had been applied including microfiltration, ultrafiltration, nanofiltration and reverse osmosis. These processes widely been used as wastewater treatment.

Ultrafiltration (UF) is effective to remove the macromolecules and the particles. However, it strongly depends on the type of material constituting the membrane [2]. Their study also found that UF membranes have been successfully used in full scale of bioreactor plant and high treatment levels landfill leachate has been achieved in such process. UF and MF are well-developed techniques used for water treatment meanwhile RO is widely used for water purification and desalination [8]. Therefore, since the success of the membrane had been proving in many applications, membrane process could be applied in treating leachate from citrus fruit. However, the performance of membrane process can be affected by several factors. Besides that, the material selection and pore size of the membrane also depends on the application it will be used. Besides, the selection of polymer membrane fabrication technique depends on the choice of polymer and desired structure of the membrane [8]. The material selection and pore size of the membrane are critical to ensuring the effectiveness of the filtration process.

II. EXPERIMENTAL PROCEDURE

There is three primary processes involved in this study as presented in Figure 1 which is conducted in the laboratory. The processes involved are leachate sampling, membrane fabrication and treatment of leachate using a membrane. The first process is the preparation and collection of citrus fruit waste leachate. The citrus fruit waste is being collected and stored until the leachate is produced. Once leachate is produced, it will be collected as for leachate sample. Then, the second process involved developed the flat sheet membrane. Meanwhile, the third process for this study is the treatment of leachate using newly fabricated flat sheet membrane. After that, several water qualities parameters will be monitored before and after the treatment using a membrane. The parameters are pH, turbidity, BOD, COD, nitrogen, phosphorus, suspended solids and colour.

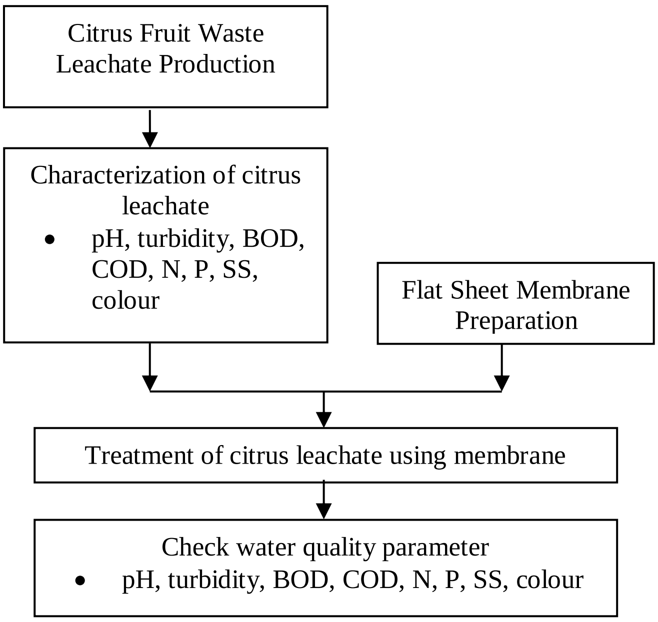

Figure 1: Flowchart of summarised methodology

A. Citrus Fruit Waste Production

The sample of leachate of this study is prepared and collected in the laboratory. The production process of leachate begins with the collection of citrus fruit waste. There are several citrus fruits were considered in this study which are lemon (*citrus limon*), sweet orange (*citrus x sinensis*), kaffir lime (*citrus hystrix*) and lime. The type of citrus fruit waste used this study is discarded citrus fruit such as damaged fruit and were collected from Pekan Besar Klang, Klang and fruit shop.The leachate reactor was also prepared as the media of citrus waste production. Then, the citrus fruit waste is stored, composted and leave to decay to form raw leachate in five plastic reactors with different weight of citrus. The leachate generated is collected every two days, and the characteristics of raw leachate were tested. Meanwhile, Table 1 shows the proportion and experimental condition of leachate sample in this study.

Table 1: Proportion of citrus fruit waste in each container

Sample	Weight of citrus fruit waste (kg)	Experimental Condition
C0	1.0	No recirculation
S1	1.0	Recirculation once every two days
S2	2.0	Recirculation once every two days
S3	3.0	Recirculation once every two days
S4	4.0	Recirculation once every two days

B. Flat Sheet Membrane Fabrication

The techniques used in this study for membrane fabrication is immersion precipitation by using membrane composition of Polysulfone (PSF) 18%, Poly (vinyl-pyrolidone)-K30 (PVP) 13% and N,N-dimenthylacetamide (DMAC) 69%.There are several steps involved in the flat sheet membrane fabrication including the preparation of dope, the membrane casting, the coagulation bath process and the solvent exchange drying process. To fabricate the flat sheet membrane, General Pneumatically Flat Sheet Membrane was used for fabrication of membrane.

C. Treatment of Leachate Using Membrane

The sample of citrus fruit waste leachate will be treated using membrane process. Before being treated with the membrane, the quality of raw leachate will be tested and recorded. Then, the sample will undergo the treatment using membrane separation process. The schematic diagram of the cross-flow filtration for the membrane separation process is as shown in Figure 2. There are several water qualities which are being monitored in this study including pH, turbidity, BOD, COD, Total Nitrogen, Phosphorus and colour

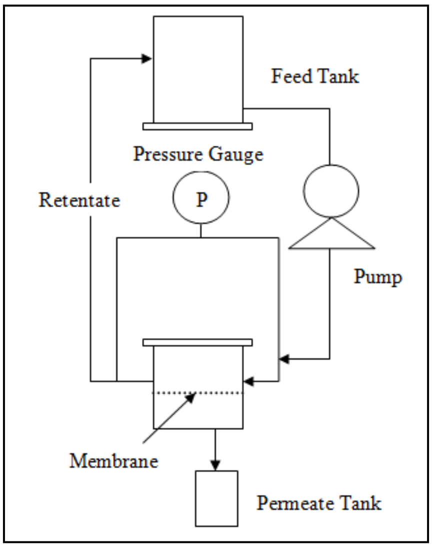

Figure 2: The Schematic Diagram of cross-flow filtration process

III. RESULT AND DISCUSSION

A. Effect of Waste Age on Quantity of Leachate Production

The volume of leachate generated gradually increased until day 5 and then experience a gradual decrease till the day of the 11 as shown in Figure 3. As for the cumulative volume in Figure 4, an S-curve graph was obtained which means that the volume of leachate generated is increasing by the day. The volume of leachate produce is increased because the citrus waste contains high moisture content that enhanced the volume of leachate generated. Besides that, the volume of leachate generated depends on the weight of waste. The higher the weight of waste, the higher the volume of leachate generated.

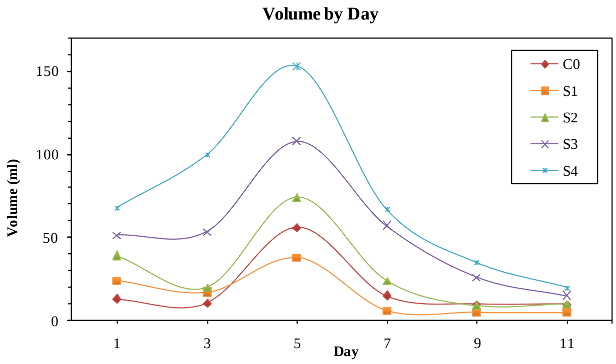

Figure 3: Volume of leachate generated by day

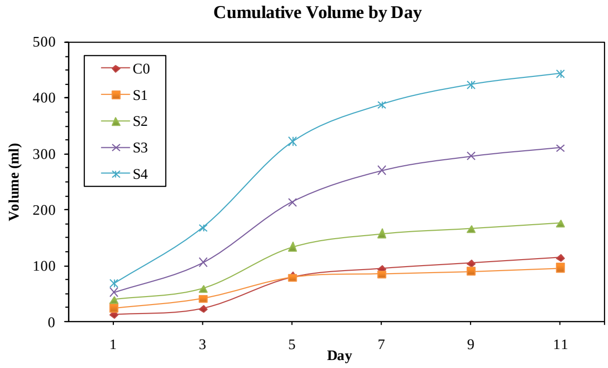

Figure 4: Cumulative volume of leachate generated

B. Effect of Waste Age on Quality Leachate Production - Physical Properties

There are three parameters of leachate quality under physical properties being monitored in this study including turbidity, total suspended solids (TSS) and colour. The turbidity of the leachate was gradually increased until day five which then slowly to decrease until the day eleventh as shown in Figure 5. From the graph, the turbidity of sample 5, S5 is the highest compared to the other samples. The turbidity of leachate is also closely related to the weight of waste. As the weight of waste increase, the values of turbidity also become higher. It is because of the more particles present in the leachate, the more light that will be scattered. Therefore, this condition caused the turbidity and total suspended solids related to each other. However, it is not a direct measurement of total suspended material in the leachate.

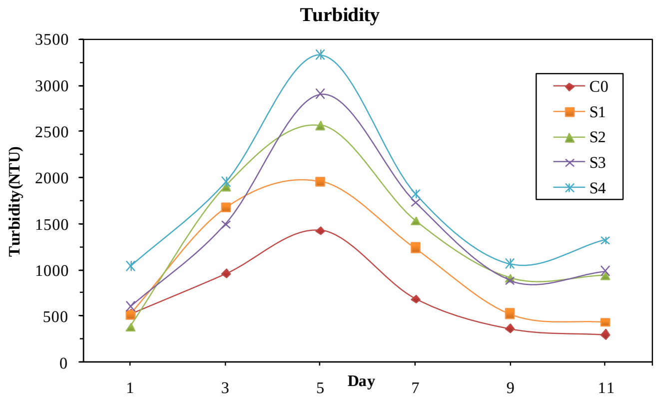

Figure 5: Turbidity of leachate

Total suspended solids (TSS) is one of the important parameter need to be taken into consideration before releasing wastewater into the water body. The allowable limit of TSS for leachate discharge by Department of Environment (DOE) is 50 mg/l for standard A and 100mg/l for standard B.

From Figure 6, the TSS of the leachate was kept increasing even though experience a little fluctuation. The amount TSS at day 11 for C0, S1, S2, S3 and S4 are 4500mg/l, 6000mg/l, 5400mg/l, 1800mg/l and 3000mg/l respectively. The amount of TSS in the citrus leachate is increasing as the waste age increase due to the decay of citrus waste particle enter the leachate and increase of microorganism. Besides that, the TSS was not influenced by the weight of waste since by following the arrangement of increased amount TSS; the highest is S2, followed by S2, C0, S4 and S3.

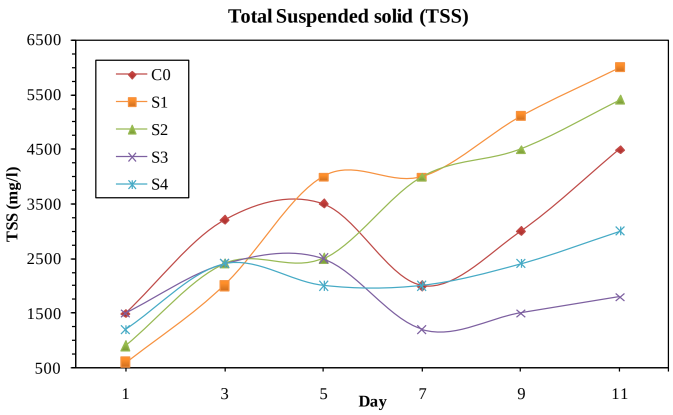

Figure 6: Total suspended solids of leachate

The colour of citrus leachate was found to be orange-brown or light brown. Associated with the leachate is a malodorous smell which is the result of the presence of organic acids, which come from the high concentration of organic matter when decomposed [9]. In their result also mention that the high concentration of colour in landfill leachate is because of the presence of the high organic substance.

Figure 7, shows the result of the colour of citrus leachate recorded for 11 days. The readings of colour for all samples are increased accordingly. In other words, the colour of the leachate is getting darker correspond to the age of leachate. This situation is supported by the study conducted by Bhalla et al. which reported that leachate produced by an old landfill with low biodegradability is classified as stabilized leachate which contains a high level of an organic substance such as humic and fluvic compound. These compounds can be indicated by the colour of leachate.

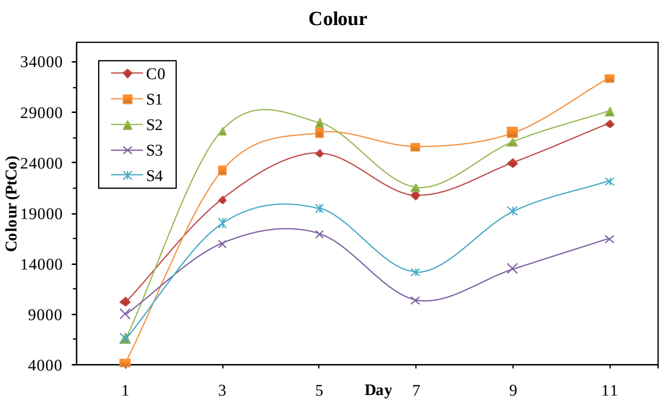

Figure 7: Colour of leachate

C. *Effect of Waste Age on Quality Leachate Production - Chemical Properties*

There are five parameters of leachate quality under chemical properties being monitored in this study including pH, Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), total nitrogen and phosphorus.

pH is classified as chemical properties which used to specify the acidity or basicity of the leachate. Figure 8, show the pH of 5 different sample of leachate with different weight of waste and experimental condition. The graph shows that the pH of leachate generated was at acidic. For sample 1, 2, 3 and 4, the pH slightly increased and decreased but remains at pH in between 3.5 to 4 which is acid. However, the control sample, C0 show a gradual increase of pH which from acidic to a neutral. CO is the leachate sample which is not undergoing the recirculation.

Bhalla et al. stated that leachate is found to have pH between 4.5 and 9. However, the pH of citrus leachate was found to be acidic and almost below 4.5. This is because the natural characteristic of citrus which is acidic. The study by Guzman et al. reported that the pH of citrus wastewater is between 3.6 to 4.5. The pH of stabilised is higher than that of young leachate [9]. Therefore, the pH of leachate is low but increased a bit because as the age of leachate increased, the ph also increased.

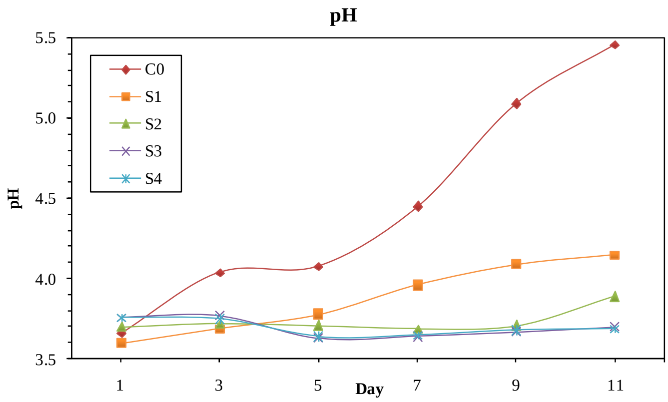

Figure 8: pH of leachate

Biochemical Oxygen Demand (BOD) is a measurement of the biodegradable organic mass of leachate, and that indicates the maturity of the landfill which usually will decrease by time. [9].

Figure 9, shows the BOD obtained in this study. The BOD of five samples for 11 days is slightly fluctuated but increase compared to the first day. The BOD of citrus leachate is high because the leachate is considered as young leachate. According to Bhalla et al., typically the BOD of landfill leachate will decrease concerning time. However, in this study, the leachate is considered young, and BOD is remaining increased by time. During day 11th, the BOD value recorded for sample C0, S1, S2, S3 and S4 are 471 mg/l, 454.2 mg/l, 458.40 mg/l, 470.40 mg/l and 472.80 mg/l respectively which is lower than the BOD of citrus wastewater reported by Guzman et al..

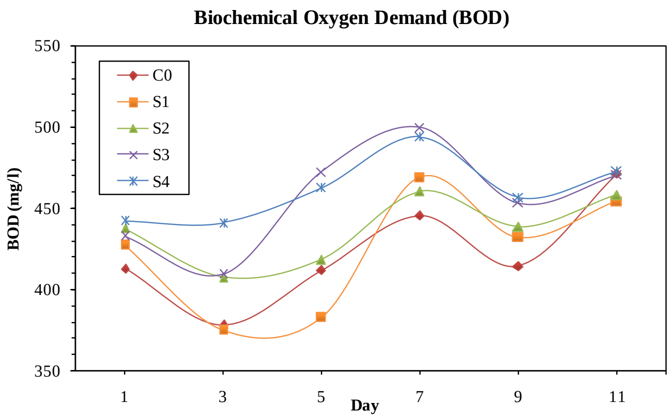

Figure 9: BOD of leachate

Chemical Oxygen Demand (COD) is the amount of oxygen required to completely oxidise the organic waste constituents chemically to inorganic end products [9].

Figure 10, shows the COD value recorded for citrus leachate within 11 days. The value of COD for five samples was high during the first day but gradually decreased for samples C0, S1 and S2 meanwhile, sample S3 and S4 experience a slight decrease but started to increase after day seventh. The COD of citrus leachate is above 25000 mg/l which is supported by the study conducted by Renou et al. [2] reported the COD value of young leachate is above 10000 mg/l. On the other hand, the COD value of citrus wastewater recorded by Guzman et al. (2016) is 10000 mg/l.

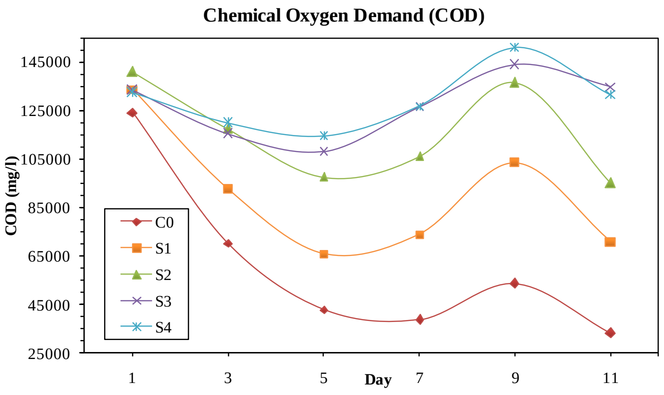

Figure 10: COD of leachate

Figure 11, show the value of total nitrogen found in the five leachate sample during 11 days. It shows that all sample experience the decreasing of TN value started day 3 until day 7. However, the value of TN increased on day 9 and day 11. The amount of nitrogen produced by leachate is corresponding to the weight of citrus waste. The value of TN is ascending from the sample of leachate with lowest weight of waste (1kg) to the highest weight of waste (4kg). From this finding, it shows that as the weight of waste increase, the volume of leachate increase, and the value of nitrogen also increase

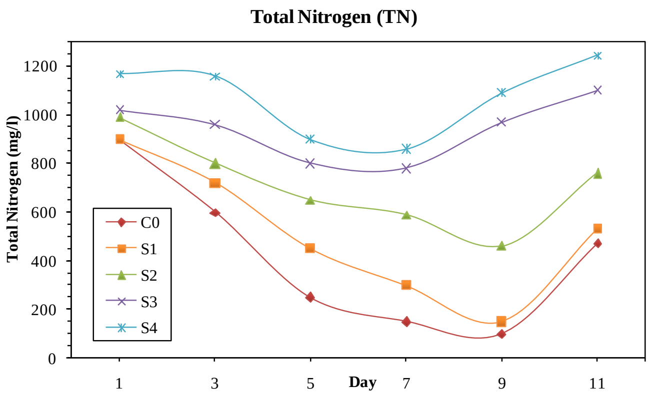

Figure 11: Total nitrogen of leachate

The major form of phosphorus that existed in the leachate was orthophosphate. (PO_4^{3-}). However, besides nitrogen, the presence of excessive amount innocuous nutrient like phosphorus can also upset the balance of aquatic ecosystem through eutrophication [12]

Figure 12, shows the generation of phosphorus in citrus leachate within 11 days. In general, the amount of phosphorus found in the leachate samples is increasing based on the weight of waste. The sample C0 and S1 which have the lowest weight of waste inherent the lowest amount of phosphorus and followed by S2, S3 and S4 which have 2kg, 3kg and 4kg of waste weight respectively. Hence, from the graph, it can be concluded that the higher the amount of citrus waste, it will increase the potential of increase in the amount of phosphorus in leachate generated.

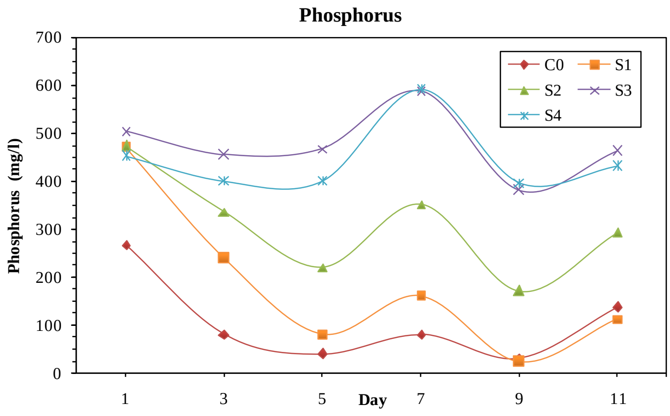

Figure 12: Phosphorus of leachate

D. *Membrane Fabrication*

The type of membrane fabricate is identified by determining the molecular weight cut-off (MWCO) of the membrane. MWCO is defined as the molecular weight of a solute that was rejected at 90%. MWCO is usually to estimate the pore size and its value can be determined from the rejection of solute of membranes against the stable molecules with various weights [11]. The non-ionized Polyethylene glycols (PEG) with different molecular weight are used to characterise the membrane fabricated in this study. Figure 13, shows a graph of rejection percentage against molecular weight. Based on the graph, the 90% rejection was fall between the molecular weight of 10 000 and 100 000 daltons. Therefore, it can be classified as Ultrafiltration which has the pore size of approximately 0.002 to 0.1 micron and an MWCO of approximately 10 000 to 100 000 daltons.

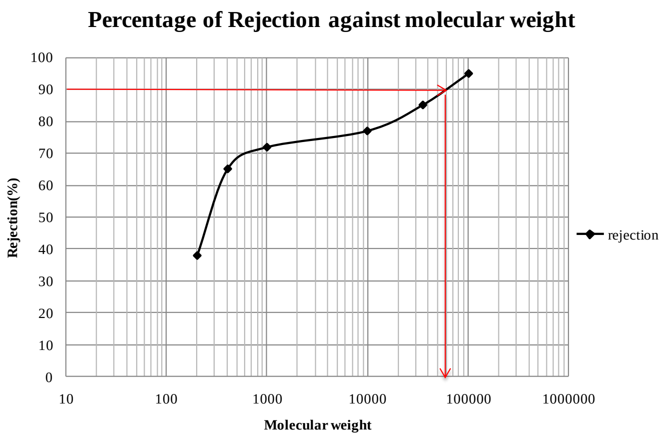

Figure 13: A graph of rejection against molecular weight

A. Treatment of Citrus Waste Leachate using Membrane

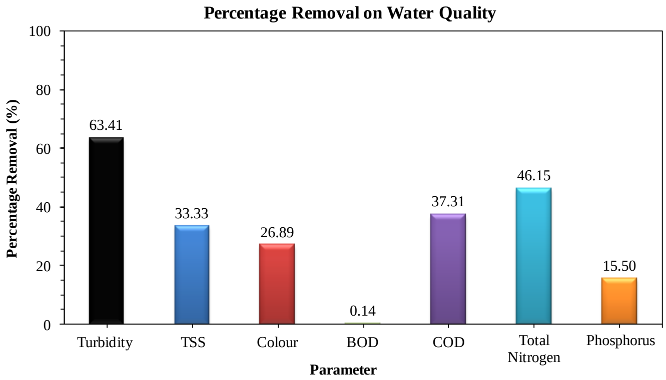

Figure 14: Percentage of removal on water quality parameters

The performance of UF membrane to treat citrus leachate is a study based on some parameters. One of the parameters is turbidity. According to Renou et al., UF is effective to remove macromolecules and particle but depending on the type of material constituent the membrane. UF might prove to be effective as a pre-treatment process for reverse osmosis and can be used to remove the larger molecular weight component of leachate which can cause fouling on reverse osmosis. In this study, UF found to be able to reduce turbidity by 63.41 %.

Another parameter to be analysed in this study is TSS. The percentage of rejection by using UF in TSS is about 33.33%. It is considered low percentage since UF has pore size approximately 0.002 to 0.1 micron which can remove larger suspended solids. However, the low percent of rejection may due to the size of the suspended particle in citrus waste is much lower than the pore size of UF. From Figure 14, the rejection of colour obtained by UF is 26.89%. This is closely related to the total suspended solids rejected by UF membrane. Suspended solids in the citrus leachate contribute to its colour. Since UF only remove about 30% of TSS, it also causes low rejection of colour since smaller size of TSS still contained in the leachate which contributes to the colour.

Besides physical properties, the chemical properties also being monitored as to analyse the performance of UF membrane to treat citrus leachate. From the study, it found that UF membrane has a limited ability to removes the pollutant in the citrus leachate. It can partially remove the contaminant in the citrus leachate. However, UF membrane in this study cannot be used to reduce the BOD since the rejection is very low which is below 1% removal. The value of BOD after treatment is higher than the permissible value which is 424.8 mg/l. The low performance of UF membrane in BOD removal may be due to the properties of the membrane such as porosity and hydrophobicity.

Chemical Oxygen Demand is one of the important factors that need to be treated before water can be released to the water body. Renou et al., reported that the elimination of polluting substance is never complete where the removal of COD is between 10 and 75%. Hence, this the reason explain the rejection of COD in the study is only 37.31%. Therefore, further treatment is needed to achieve the standard limit of discharge by DOE which is 400 mg/l. The percentage of nitrogen removal is the second highest after turbidity which is 46.15%. However, the amount of TN after treatment unable to meet the requirement of standard limit by DOE and consider high which can cause eutrophication

From the Figure 14, the percentage of phosphorus removal is 15.50%. The percentage is lower than the percentage removal of TN by UF membrane. There is the possibility of the particle size of phosphorus is larger than nitrogen particle. The permissible phosphorus value by DOE is 0.2 mg/l is far from the value of phosphorus even after the treatment which is 169 mg/l.

IV. CONCLUSION

Based on the findings of this study, we can conclude that the weight of citrus waste influences the volume of leachate generated. The volume of leachate will increase but then gradually decreased by time. As higher the weight of waste, the higher the volume of leachate will be generated. The waste age also gives effect to the quality of leachate generated. The quality of leachate various depend on the waste age and also the weight of waste. The quality of citrus leachate has a high potential to pollute the environment because this type of leachate characterised by very high organic loads which are BOD above 400 mg/l and COD above 3000 mg/l. Besides that, citrus leachate has low pH which is acidic. This condition is not favourable because, in conventional treatment, pH must fall between 6.0 to 8.5 to accelerate the biological processes and to ensure the occurrence of sedimentation of the mud.

Other than that, the presence of nutrient which is Nitrogen and Phosphorus in citrus wastewater is considered high. Therefore, immediate treatment is needed since these types of nutrient contributed to the eutrophication. Eutrophication is the situation occurs due to the excessive nutrient such as nitrogen and phosphorus in the water body which can cause a dense growth of plant life and death of animal life resulting from lack of oxygen. The UF membrane fabricated in this study is not enough to reduce the pollutants contains in the citrus leachate. The composition of membrane might be the reasons for the low performance of membrane process. The pore sizes of the membrane also affect its performance. From this study, with pore size of UF membrane is insufficient to treat citrus leachate

As the conclusion, the characteristic of citrus leachate is far exceeding the permissible value of discharge set by DOE. Therefore, it needs to treat due to its potential to pollute the environment. It also found UF membrane insufficient to treat citrus leachate.

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REFERENCES

[1] Ahmed, F. N., & Lan, C. Q. (2012). Treatment of Landfill Leachate using Membrane Bioreactors: A review. *Desalination* , 41-54.

[2] Renou, S., Givaudan, J. G., F., D., & Moulin, P. (2008). Landfill Leachate Treatment: Review and Opportunity. *Journal of Hazardous Materials* , 468-493.

[3] Johari, A., Ahmed, S. I., Hashim, H., Alkali, H., & Mat, R. (2012). Economic and Environmental Benefits of Landfill Gas from Municial Solid Waste in Malaysia. *Renewable and Sustainable Energy Reviews* , 2907-2912.

[4] Thi, N. B., Kumar, G., & Lin, C.-Y. (2015). An Overview of Food Waste Management in Developing Countries: Current Status and Future Perspective. *Journal of Environmental Management* , 220-229.

[5] Lam, S. S., Liew, R. K., Lim, X. Y., Ani, F. N., & Jusoh, A. (2016). Fruit waste as feedstock for recovery by pyrolysis technique. *International Biodeterioration & Biodegradation* , 325-333.

[6] Garcia, E., Gozalvez, J. M., & Lora, J. (2002). Use of Reverse Osmosis as a Preconcentration System of Waste Leaching Liquid from the Citric Juice Production Industry. *Desalination* , 137-142.

[7] Saverini, M., Catanzaro, I., Sciandrello, G., Avellone, G., Indelicato, S., Marci, G., et al. (2012). Genotoxicity of Citrus Wastewater in Prokaryotic and Eukaryotic Cells and Efficiency of Heterogenous photocatalysis by Tio2. *Journal of Photochemistry and Photobiology B: Biology* , 8-15.

[8] Laila, S. B., Kochkodan, V., Hashaikeh, R., & Hilal, N. (213). A review on membrane fabrication: Structure, properties and performance relationship. *Desalination* , 77-95.

[9] Bhalla, B., Saini, M. S., & Jha, M. K. (2013). Effect of Age and Seasonal Variations on Leachate Characteristics of Municipal Solid Waste Landfill. *International Journal of Research in Engineering and Technology , 2* (8), 223-229.

[10] Guzman, J., Mosteo, R., Sarasa, J., Alba, J. A., & Ovelleiro, J. L. (2016). Evaluation of Solar Photo-Fenton and Ozone Based Process as Citrus Wastewater Pre- treatments. *Separation and Purification Technology* , 155-162.

[11] Hamzah, S., Ali, N., Mohd Ariffin, M., Ali, A., & Mohammad, A. W. (2014). High Performance of Polysulfone Ultrafiltration Membrane: Effect of Polymer Concentration. *Journal of Engineering and Applied Science , 9* (12), 2543- 2549.

[12] Praveen, P., & Loh, K.-C. (2016). Nitrogen and Phosphorus Removal from Tertiery Wastewater in an Osmotic Membrane Photobioreactor. *Bioresource Technology* , 180-187.

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