Onsite treatment of higher-load graywater by membrane bioreactor

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Introduction

Water resources had decreased their qualities through the years and clean water is expected to be a scarce resource in the near future. In this regard, Onsite Wastewater Differentiable Treatment System (OWDTS) is proposed to answer the problems on water issues. In this concept, wastewater from a household is fractioned into three and will be treated separately. These are blackwater (feces and urine), higher-load graywater (kitchen sink and washing machine wastewaters) and lower-load graywater (shower, bath, and wash basin wastewaters) (Lopez Zavala *et al*., 2002).

Higher-load graywater (HLGW) is the mixture of kitchen sink wastewater (KSWW) and washing machine wastewater (WMWW) and among the five graywater discharges from the household, these two have high percentages of contribution to the pollution load in terms of chemical oxygen demand (COD), nitrogen (N), and phosphorus (P) components (Almeida *et al*., 1999; Eriksson *et al*., 2002). Furthermore, WMWW contains high concentration of the surfactants that are present in the detergent used in washing clothes. Linear alkylbenzene sulphonates (LAS) is the most important surfactant used and is considered as a workhorse surfactant in both powder and liquid laundry products (de Guertechin, 1999). Researchers (Patterson *et al.*, 2001; Abu-Hassan *et al*., 2006) have shown that biodegradation of many surfactants, including LAS, may be restricted between concentrations of 20-50 mgL^{-1} and may be inhibited at higher concentration. Therefore, treatment of HLGW and monitoring its LAS concentration is necessary before it is being discharged to the environment or reused for another purpose.

Membrane bioreactor (MBR) has been recently applied to treatment of domestic wastewater and other types of graywater discharges. In this paper, the MBR was applied for onsite treatment of HLGW and was operated at constant transmembrane pressure (TMP), thus no pump requirement for permeation. Furthermore, the MBR was modified such that it could accommodate the high fluctuations of the wastewater that enters into the system throughout the day without the need of the equalization tank and pumping system. In this regard, a simple and less energy consuming system was introduced.

The primary objective of this research was to investigate the application of subMBR as possible technology in the treatment of HLGW. The detailed objectives were:

- To determine the effect of organic loading rate (OLR) on the treatment of the following type of wastewaters using ultrafilter hollow fiber membrane:
  - KSWW only
  - HLGW mixture
- To determine the effect of the following operation styles on the treatment of HLGW using microfilter flatplate membrane:
  - Continuous feeding operation
  - Intermittent feeding operation
- To determine the fate of LAS
- To compare with the Johkasou system

Materials and Methods

Membrane bioreactor and wastewater samples for determining the effect of OLR

The membrane bioreactor used in this study is schematically described in Fig. 1. A hollow fiber (HF) membrane configuration was used. It was made of poly acryl nitrile with a pore size of 100 kDa (ultrafilter). Four lab-scale MBR systems were operated at different hydraulic retention times (HRT) to give different OLR. The first set of these 4 MBRs was used to treat KSWW only; the next set was used to treat the HLGW mixture (1:1 ratio of KSWW and WMWW). Activated sludge was obtained from Shinkawa Wastewater Treatment Plant in Sapporo, Japan. The kitchen sink wastewater supplied daily was obtained during its peak hours (2 pm, after washing dishes from lunch) from the cafeteria of Faculty of Engineering, Hokkaido University. It was passed through a screen and oil was removed before storing in the refrigerator and continuously supplying to the system. On the other hand, WMWW was obtained from students at the same faculty. Sludge was withdrawn regularly to maintain an MLSS of 11-13 gL^{-1}. The average airflow rate was 2.5 $Lmin^{-1}$ in each reactor.

Samples from the influent, inside the reactor, and permeate were obtained for analyses of the following parameters: COD, TKN, NO_x-N, TP, PO_4-P, and LAS. Membrane flux, MLSS and molecular weight distribution (MWD) in terms of DOC were also monitored.

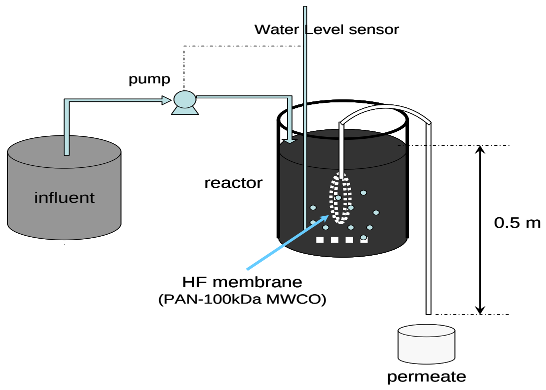


Fig. 1 Configurations of a single **subMBR**

Continuous feeding operation

The membrane bioreactor applied in this study employed a microfilter flat-plate membrane system (pore size of 0.4 μm, area = 0.1 m^2, polyolefin) from Kubota Company. The reactor has the following dimensions: 630 mm x 90 mm x 227 mm (H x W x L). The effective volume of 10L was used (see Fig. 2a).

The activated sludge was obtained at Shinkawa Wastewater Treatment Plant in Sapporo, Japan. Since the 1:1 mixture of the two wastewaters was continuously supplied to the system, an equalization tank was necessary. A synthetic graywater was used (Huelgas, 2009). Permeate was intermittently (10 min on-2 min off) withdrawn at a certain TMP. The TMP of 1.5 kPa was set for the first 12 days and increased gradually to 3.0 kPa until day 14^{th}. TMP of 3 kPa was maintained for the remaining of the operation. A water level sensor was used to send signal to the peristaltic pump to supply raw graywater stored in a refrigerator, thereby maintaining the volume of the mixed liquor inside the reactor. Air compressor and diffuser were used to supply air to the system at a flow rate of 10 L min^{-1} and the dissolved oxygen concentration was monitored and maintained above 4 mgL^{-1}. Mixed liquor temperature was maintained at 20^{o}C by a water bath. Samples from the influent, reactor, and permeate were obtained for subsequent analyses.

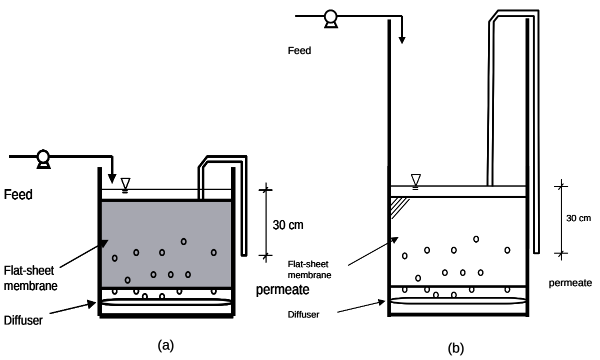


Fig. 2 Configuration of the MBR system for the (a) continuous feeding operation (b) intermittent feeding operation

Intermittent feeding operation

The MBR applied in this study is basically the same as that in continuous-feeding operation (Fig. 2b). However, the reactor size was modified such that the influx of WW in the morning and the evening will be accommodated without the use of an equalization tank. The MBR has a space to receive this influx of WW during its peak hour discharges (Huelgas 2009). The influent was supplied twice a day. A programmed sequencer was used to automatically supply the raw wastewater into the MBR. Ten liters (10 L) of the 1:1 HLGW mixture was discharged evenly from 7:00 to 8:30 am in the morning, therefore 1L of graywater was discharged at 9 minutes interval. In the evening, 1L of KSWW is discharged every 18 minutes from 19:00 to 22:00 giving a total of 10 L. Permeate was intermittently (10 min on-2 min off) withdrawn at a constant minimum TMP induced by a water level difference between the reactor and the permeate. TMP and flux varied throughout the day and the hourly and weekly variations were monitored. Air compressor and diffuser were used to supply air to the system at a flow rate of 10 L min^{-1}.

Samples in the influent, inside the reactor, and permeate were obtained for analyses of the following parameters: COD, TKN, NO_x-N, TP, PO_4-P, and LAS. Membrane flux and MLSS were also monitored.

Results and Discussion

Effect of organic loading rate

The effect of OLR in the treatment of KSWW only and HLGW mixture was investigated. The influent COD of the KSWW and the HLGW mixture were 1300 mgL^{-1} and 890 mgL^{-1}, respectively. These are higher than the domestic raw wastewater with a range of 250-800 mgl^{-1} (Tchobanoglous *et al*., 2003), and the total graywater discharge with a range of 495-682 mgl^{-1} (Palmquist and Hanæus, 2005).

Table 1 shows the OLR, MLSS, and F/M ratio for each corresponding HRT in both series of experiments. The higher value of OLR in the KSWW (6.9 $kg_{COD}m^{-3}d^{-1}$) compared to that of the mixture (5.3 $kg_{COD}m^{-3}d^{-1}$) at HRT of 4 hours is due to the high COD value of the KSWW influent than that of the mixture. The same trend was observed at all other HRTs. Furthermore, the F/M ratios were relatively higher for MBR systems treating the KSWW only compared to its corresponding reactors treating the mixture. And in both cases, Reactor 1 has the highest F/M ratio among Reactors 1, 2, 3, and 4. It has been reported that F/M ratios for MBR systems are <0.2 $kg_{COD}kg_{MLSS}^{-1}d^{-1}$ and even approach 0 at long SRT and high sludge concentration (Stephenson *et al*., 2000).

Reactor 1 treating the KSWW has higher OLR values than when treating the HLGW mixture, but foaming was observed in the system treating the mixture which was the reason why the reactor was stopped after few days of operation.

Table 1 Operating condition of the reactors in the treatment of KSWW and HLGW

Reactor	HRT hr	OLR $kgm^{-3}d^{-1}$	MLSS gL^{-1}	F/M ratio $kg_{COD}kg_{MLSS}^{-1}d^{-1}$
KSWW				
R1	4.5	6.9	11-13	0.58
R2	7	4.5	11-13	0.38
R3	12	2.6	11-13	0.22
R4	24	1.3	7-9	0.16
KSWW +WMWW				
R1	4	5.3	9-11	0.53
R2	8	2.7	9-11	0.27
R3	12	1.8	9-11	0.18
R4	24	0.9	6-7	0.14

Table 2 summarizes the characteristics of the permeate. In the treatment of the HLGW mixture, the reactor with longest HRT (24 hours) has the lowest COD range of 12-51 mgL^{-1} (average COD = 29.3 mgL^{-1}). Reactors 2 and 3 have average COD of 82.5 mgL^{-1} and 59.4 mgL^{-1}, respectively. These values are higher compared to the system operated at almost the same condition but treating KSWW only with an average COD of 29, 17 and 17 mgL^{-1} for HRT of 7, 12, and 24 hours, respectively (Huelgas, 2006). The COD is almost double in the system treating the mixture with that system treating KSWW only. This may indicate that some part of the WMWW may not be easily biodegradable.

Table 2 Permeate quality of the reactors in the treatment of KSWW and HLGW

Reactor	COD mgL^{-1}	NH_4-N mgL^{-1}	NO_3-N mgL^{-1}	PO_3-P mgL^{-1}
KSWW				
R1	35-73 (53)	-	-	-
R2	11-53 (29)	-	-	0.1
R3	10-26 (17)	-	-	0.8
R4	7-34 (17)	-	13	2.4
KSWW +WMWW				
R1	-	-	-	-
R2	48-123 (82.5)	0.1	0-0.01	-
R3	28-124 (59.4	0.55	0-0.41	-
R4	12-51 (29.3)	0.06	0-3.67	0-0.54

In Japan, washing machine discharges three sets of wastewater during one cycle of washing clothes. Most of the pollutants including surfactants (like LAS) can be found in the first discharge of the washing machine. The measured total LAS concentration in the first discharge was in the range of 20.59 – 46.24 mgL^{-1} with an average value of 35.06 mgL^{-1}. Since the influent used in this study was a mixture (1:1) of KSWW and WMWW, the LAS concentration in the influent is around 17.53mgL^{-1}. This value is relatively higher than the reported concentration values of LAS in the domestic wastewater which is on the range of 1-15 mgL^{-1} (Zoller, 2004). This is because the main source of LAS is WMWW and this is more concentrated in the HLGW.

In the treatment of the HLGW mixture, the range of the total LAS concentration in the permeates of reactors 2, 3 and 4 were 37- 2341 μgL^{-1}, 11-2457 μgL^{-1}, 8-502 μgL^{-1}, respectively (Fig. 3). The lower value in reactor 4 was due to low loading rate and longer sludge retention time which enhances biodegradation of this micro pollutant. Regardless of HRT or loading rate, very high removal rate of LAS was obtained, even up to > 99%. Same removal rate has been observed in other papers with domestic wastewater influent (Temmink, 2004; De Wever, 2004). This high rate of removal can be accounted also to the characteristics of the MBR systems which include complete retention of solids among others. This indicates that there is no inhibition in the biodegradation of LAS at the range of influent concentration around 10.3-23.1 mgL^{-1}.

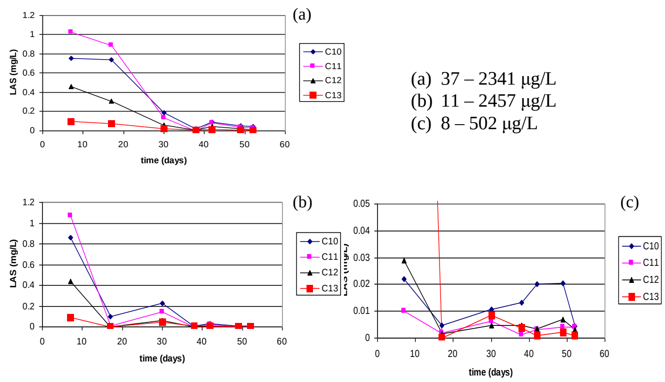


Fig. 3 LAS concentration in the permeate at (a) Reactor 2, (b) Reactor 3, and (c) Reactor 4

From all these results, we can conclude that the MBR can be operated at HRT of 8 hours and longer. It corresponds to an OLR of 4.6 $kg_{COD}m^{-3}d^{-1}$ and 2.7 $kg_{COD}m^{-3}d^{-1}$ for KSWW and HLGW mixture, respectively. And the COD of the permeate of the system treating the mixture is higher than the system treating the KSWW only.

The high removal rate of LAS supports that the remaining organic matter (OM) is not LAS in its original form. The nitrates and phosphates were of low concentrations, confirming that they are not abundant in graywater but rather in blackwater. Low flux was observed (Fig. 4) which gave another option to use another membrane, a micro-filter, flat plate (MF-FP) membrane in the succeeding experiments.

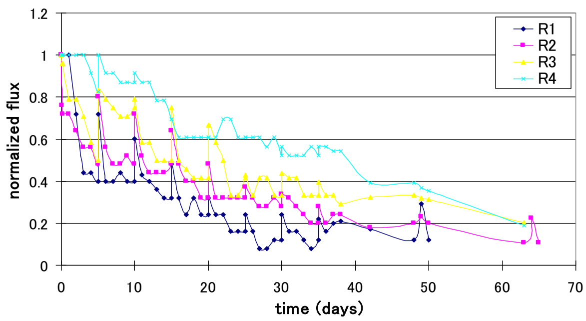


Fig. 4 Flux through time during the treatment of KSWW only

Effect of continuous and intermittent feeding operation

Continuous feeding operation

The lab-scale MBR was continuously operated for 87 days. The HRT was at around 10 hours which increased to 16 hours towards the end of the operation. The average HRT during the whole duration was 13.6 hours. This can be attributed to the decrease in membrane flux. The MBR has an average flux of 0.22 $m^3m^{-2}d^{-1}$ (Fig. 5). The flux decreased from 0.28 $m^3m^{-2}d^{-1}$ to 0.18 $m^3m^{-2}d^{-1}$. Some studies on MBR treating municipal and domestic wastewater report membrane flux values between 0.12-0.96$m^3m^{-2}d^{-1}$ (Stephenson *et al*., 2000). The low flux observed in the present system was due to the operation at constant TMP. The TMP used was also low compared with those observed in the system operated at constant flux. Increasing the constant TMP applied is expected to increase the flux.

MBR system has an advantage of dealing with longer SRT (or even complete retention of sludge) and a high MLSS concentration. The MLSS concentration in this experiment ranged from 10-25 gL^{-1}. The average OLR and F/M ratio were 1.21 $kg_{COD}m^{-3}d^{-1}$ and 0.07 $kg_{COD}kg_{MLSS}{}^{-1}d^{-1}$, respectively. The F/M ratio was lower compared to the previous experiment (Table 1) treating the mixture because no sludge withdrawal was done in this experiment resulting to the MLSS concentration reaching to as high 25 gL^{-1}.

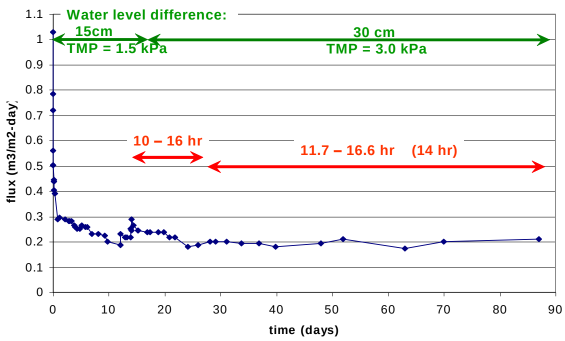
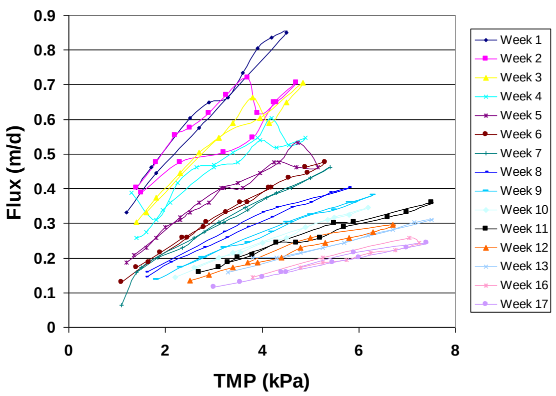


Fig. 5 Flux through time during the continuous feeding operation

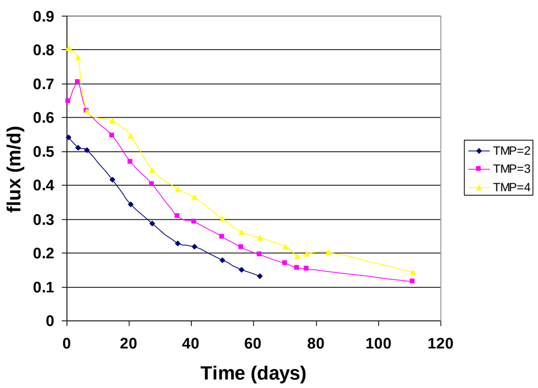
The qualities of the influent in terms of COD, N, and P were measured and the HLGW mixture has a total COD of 675 mgL^{-1}. This is lower compared to that obtained from the real wastewater sample which has a total COD of 890 mgL^{-1}. Although the KSWW was simulated to give the same COD value as that of the real KSWW samples, the WMWW's COD is lower than that of the real samples because the clothes used for washing are basically clean. Therefore, the contribution of pollutants coming from the used clothes is not considered in this experiment. Furthermore, the total LAS concentration in the influent was measured at around 30.8 mgL^{-1}. This concentration is higher than that observed in municipal wastewater plants dealing only with domestic wastewater which has a range of 1-15 mgL^{-1} (Zoller, 2004). This is because the main source of LAS is WMWW and this is more concentrated in the HLGW. However, it was furthermore observed that this value is also higher than that obtained from the real WMWW samples which has an average concentration of 17.53 mgL^{-1}.

Intermittent feeding operation

The reactor was continuously operated for 120 days. The membrane flux was measured throughout the day as the TMP changes due to the intermittent supply of influent. Fig. 6a shows the flux against TMP for the morning discharge and Fig. 6b shows the flux plotted against time at TMP of 2, 3, and 4 kPa. A decrease in flux was observed as the weeks proceeded reaching 0.12 md^{-1} at TMP of 3 kPa. It was observed that the system continuously treating 1:1 ratio of KSWW and WMWW has a constant flux of 0.2 md^{-1} at TMP of 3 kPa (Fig. 5). At TMP of 3 kPa, the intermittent feeding operation has an advantage of higher flux until 60 days of operation.



(a)



(b)

Fig. 6 (a) Weekly measurement of flux against TMP variations (b) Flux decline through time at TMP of 3, 4, and 5 kPa (in the morning discharge)

The COD of the HLGW mixture in the morning and the KSWW only in the evening were 675 mgL^{-1} and 1050 mgL^{-1}, respectively. The MLSS concentration was maintained at 16 gL^{-1}. Variations in the quality of treated wastewater in terms of COD, N and P are measured. Fig. 7 shows the COD of the permeate from the composite samples of the morning and evening discharges. It showed that the COD of the permeate from the morning discharge was higher than that of the evening discharge regardless of the fact that the influent COD of the morning discharge is smaller than that of the evening discharge. This has been observed also in previous experiments (Table 2) wherein the permeate obtained from the treatment of the HLGW mixture is higher than that of the permeate obtained from the treatment of KSWW only.

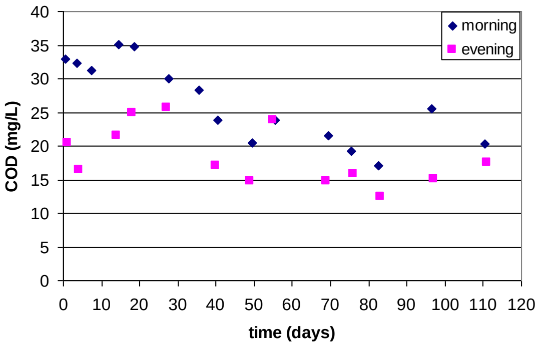


Fig. 6 COD of the permeate from the composite samples of the morning and evening discharges

Fate of LAS

Previous results (Table 2) showed that organic matter (in terms of COD) in the permeate of the HLGW mixture was higher than that of the KSWW regardless of the fact that the HLGW mixture has a lower influent COD concentration compared to KSWW. This implies that some components in the mixture are not completely degraded. However, the parameters that affect this cannot be easily pointed out because both systems are not totally subjected to the same parameters. Therefore a batch experiment was performed subjecting the two types of wastewater into the same operating conditions to confirm that it is the characteristics of the WW that influence this result and not the differences in the operating conditions. Also, the component that gives the permeate of the HLGW mixture higher COD than that of KSWW was determined.

It has been found that through time, the organic matter in terms of DOC in the mixture was higher than that of the KSWW only as shown in Fig. 7. This result implies that WMWW has some components that are not easily biodegradable. Furthermore, the influent LAS concentration can be degraded up to > 99% in all the experiments. Therefore, the remaining OM is not LAS in its original form.

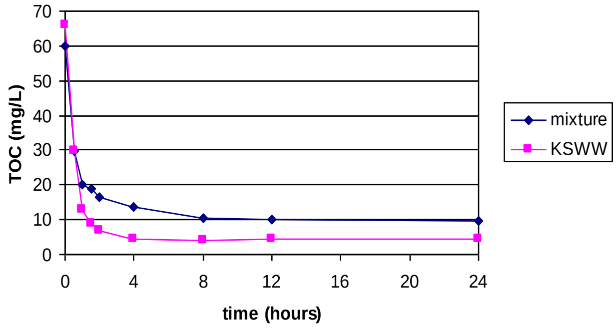
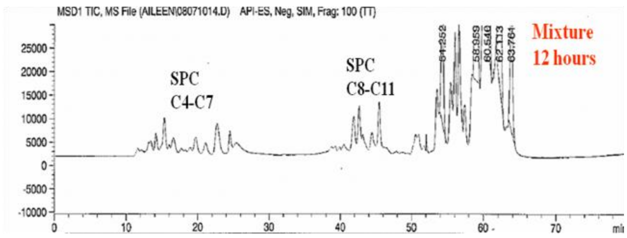
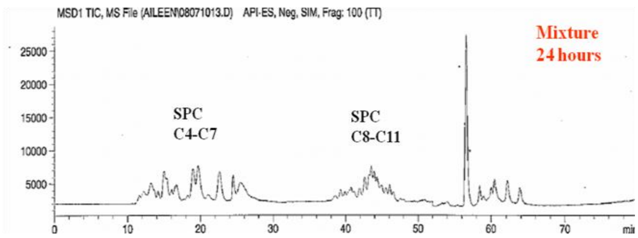


Fig. 7 DOC profile through time



(a)



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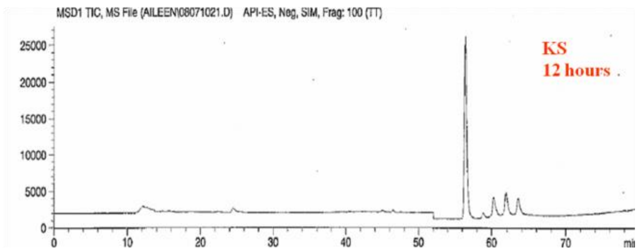
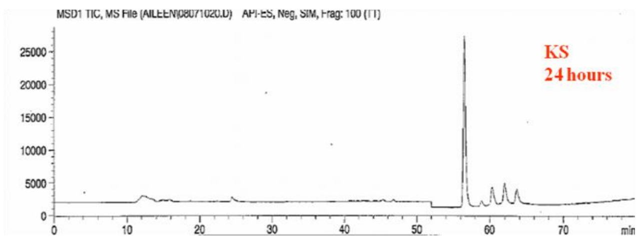


(c)



(d)

Fig.8 LC/MS chromatogram for the following: HLGW mixture after (a) 12 hr; (b) 24 hr; KSWW only after (c) 12 hr; (d) 24 hr

The samples were analyzed for the degradation of LAS and the formation of its by-products, the SPC. LAS has been removed but was not completely degraded into CO_2 and H_2O. The LC-MS chromatogram at SIM mode showed that SPCs were detected even after 12 hours and 24 hours of operation for the HLGW mixture, as shown in Fig. 8 (a) and (b), respectively. This must be the reason why the COD or DOC of the mixture is higher than that of the KSWW only. Fig. 8 (c) and (d) show the absence of these substances in the system treating KSWW only after 12 hours and 24 hours of operation. However, the amount of SPC cannot be quantified due to lack of standard solution.

Comparison with Johkasou system

The performance of the three types of Johkasou systems: the anaerobic filter-contact aeration, anaerobic filter-moving bed biofilm, and the membrane Johkasou were discussed by Lens *et al.,* 2001. The total tank volume for anaerobic filter-contact aeration, anaerobic filter-moving bed biofilm, and membrane Johkasou are 3.55 m^3, 3 m^3 and 2.488 m^3, respectively. The HLGW treatment using a continuous operation mode requires an equalization tank which volume was set to accommodate a whole day's HLGW discharge. The equalization tank's volume is 0.187 m^3 in addition to the main reactor's volume which is 0.0936 m^3 giving a total volume is 0.28 m^3. On the other hand, the volume of the subMBR using an intermittent operation mode is 0.187 m^3 which is the sum of the main reactor's effective volume of 0.0936 m^3 and the buffer tank's volume of 0.0936 m^3 (Huelgas, 2009).

The volume of the composting toilet was based on the commercially available "Bio-Lux" (Model: S-15). The main body volume of the composting toilet is 0.38628 m^3 (1 m x 0.620 m x 0.623 m). This was added to the volume of the MBR for the treatment of HLGW to get the total volume for the treatment using the concept of source separation. Therefore, in the OWDTS system, the combination in terms of volume is still smaller compared to that of Johkasou systems by four times. However, the maintenance being applied to the Johkasou systems should also be considered for the treatment of the HLGW using MBR.

Conclusions and Recommendations

Onsite treatment of higher-load graywater using subMBR has been investigated. The following has been determined: (a) the effect of organic loading rate on the treatment of the treatment of KSWW only and HLGW mixture using UF-HF membrane; (b) the effect of the continuous feeding operation and intermittent feeding operation styles on the treatment of HLGW mixture using MF-FP membrane; (c) the fate of LAS, and (d) the comparison with Johkasou membrane.

It has been found that the MBR can be operated at HRT of 8 hours and longer for both the treatment of KSWW only and the HLGW mixture. Higher organic matter in the permeate of the system treating the mixture was obtained compared to the system treating KSWW only, implying that WMWW has some components that are not easily biodegradable. The high removal rate of LAS (> 99%) indicated that the remaining OM is not LAS in its original form. The nitrates and phosphates were of low concentrations confirming that they are not abundant in graywater but in blackwater. Low flux was observed which gave another option to use a micro-filter, flat plate (MF-FP) membrane in the succeeding experiments.

Two types of operation style have been investigated: the continuous feeding and the intermittent feeding type of the operation. The continuous feeding gave a stable membrane flux throughout the operation but the intermittent feeding operation exhibited better membrane flux performance up to a certain time. During the first 60 days, the MBR operated with continuous feeding gave a lower flux at TMP of 3 kPa compared to that operated with intermittent feeding. It has been observed also that the composite sample from the morning discharge has a higher COD than that of the evening discharge regardless of the fact that the influent COD of the latter is higher. This can be accounted to the presence of biodegradation intermediate by-products of LAS which is SPC. The lack of standard solution for the SPC made it difficult for its quantification. It is therefore recommended that this can be quantified to determine the amount it contributes to the COD in the permeate
Comparison with the Johkasou system showed that there is around four times reduction in the volume of the treatment facility if source separation is considered.

It is recommended that the results of these experiments will be applied to assess the economical aspect of onsite graywater treatment using MBR systems.

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