

THE USE OF A SUBMERGED MEMBRANE BIOREACTOR FOR THE TREATMENT OF A STYRENE CONTAINING SYNTHETIC WASTEWATER

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ABSTRACT

In this study, the potential of a membrane bioreactor for treating styrene contaminated synthetic wastewater, with COD that simulated a petrochemical wastewater, was evaluated and compared with previous studies. At hydraulic retention times of 18 and 24 hours, the removal efficiency of COD and styrene was consistently higher than 99%. More significantly no styrene was detected in the exhaust air, which meant that, at both hydraulic retention times, the mechanism of styrene removal in the membrane bioreactor was exclusively through biodegradation. The lower contribution of stripping to overall styrene removal obtained in the present work compared to that previously reported for traditional activated sludge processes was attributed to the higher Mixed Liquor Suspended Solids (MLSS) that can be achieved in a membrane bioreactor. Decrease of hydraulic retention time from 24 h to 18 h also resulted in significant increase in the rate of membrane fouling as quantified by transmembrane pressure gradient. This was attributed to decrease in extracellular polymeric substance concentration which resulted in sludge deflocculation. As a consequence, the particle size distribution of the activated sludge shifted to lower mean particle sizes and the increase in the percentage of smaller particles resulted in increase in the rate of membrane fouling.

Key words: Activated sludge, Biodegradation, Immersed membrane bioreactor, Styrene, Volatile organic compound

INTRODUCTION

The most common method employed for degradation of hazardous volatile organic compound (VOC) chemicals such as styrene in petrochemical wastewaters is the conventional activated sludge process (CASP). The treatment of petrochemical wastewaters using CASP usually results in effluents which meet the industrial standards for the disposal of wastewaters (Qin *et al.*, 2007). However, the CASP have two major disadvantages. The first is that during the aeration of the activated sludge, a significant quantity of VOCs can transfer to the air phase exiting these processes (Hsieh, 2000; Lin *et al.*, 2004; Cheng *et al.*, 2008). Since styrene has been classified as

a potential human carcinogen and occupational exposure limits have been set for it (USEPA, 1994), this means that the gaseous effluent from activated sludge plants requires further treatment. The second disadvantage is that the effluent from these units does not usually have the standard necessary to enable its use as process water (Qin *et al.*, 2007).

According to the paper by Hsieh (2000), the removal of VOCs from industrial wastewaters during CASP occurs by the three mechanisms of stripping, adsorption and biodegradation. According to the model developed in this paper, the percentage contribution of biodegradation to overall styrene removal can be increased through the increase of either Mixed Liquor Suspended

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Solids (MLSS) or the biological rate constant for biodegradation. The latter can be achieved through the use of activated sludge previously adapted to styrene, whereas the former can be attained through the use of novel processes such as membrane bioreactor (MBR).

MBR is an activated sludge process in which the sedimentation stage has been replaced with membrane filtration. In this way, both a high MLSS and a very good quality effluent, suitable for process applications or may be even drinking water, can be achieved (Judd, 2006; Qin *et al.*, 2007; Tam *et al.*, 2007). The concentration of MLSS in MBRs is reported to be typically 3-5 times that of CASPs (Qin *et al.*, 2007).

There have been several reports on the application of MBR process to the treatment of petrochemical wastewaters. Qin *et al.* (2007) claimed that a MBR process with an hydraulic retention time (HRT) as low as 13 hours can be used to treat a petrochemical wastewater and to obtain an effluent that meets discharge standard. Chang *et al.* (2006 a) claimed that BOD and COD removal efficiencies obtained with a MBR process used for the treatment of Acrylonitrile-Butadiene-styrene (ABS) industrial wastewater is higher than that which could be achieved with a CASP.

A drawback of the use of MBRs, compared to that of CASPs, is the possibility of membrane fouling, which, through decrease of the membrane flux, can severely reduce the output from such processes (Judd, 2006). Previous works have shown that the membrane fouling is affected by factors such as HRT, sludge retention time and biomass characteristics such as floc diameter, morphology, extracellular polymeric substances (EPS), soluble microbial product (SMP), and viscosity (Ng and Hermanowicz, 2005; Massae *et al.*, 2006; Meng *et al.*, 2007). Therefore, the increase in MLSS, which is necessary to reduce the contribution of stripping to VOC removal in a CASP (Hsieh, 2000), can also aggravate the membrane fouling in a MBR.

A common method of alleviating membrane fouling in both submerged and crossflow MBRs (Ndinisa *et al.*, 2006; Psoch and Schiewer, 2006; Meng *et al.*, 2008) is air scouring of the membrane; this means that in MBRs, air sparging has the dual roles of supplying the mixed bacterial culture

in activated sludge with oxygen and reducing the formation of the foulants on the membrane. For this reason, the air rates reportedly used in MBRs processes is usually higher than that used in CASPs. However, according to the model of CASP developed by Hsieh (2000), increase in air rate in CASP can result in significant increase in the rate at which VOCs, such as styrene, are removed by the stripping mechanism.

In the present study the performance of MBR in terms of COD and styrene removal and stripping of styrene was studied and compared with findings of previous work with the same or similar wastewaters in MBR and traditional activated sludge processes. Additionally, the membrane fouling during the operation of the MBR was characterized.

MATERIALS AND METHODS

Seeding organisms

Activated sludge, used as inoculum in MBR runs, was obtained from the sludge return line at the CASP of Tabriz petrochemical company. After transfer to the laboratory, the activated sludge was frozen using 15% w/w glycerol and was stored. Before inoculating the MBR, the frozen activated sludge was thawed and grown under aeration in the above-mentioned media without styrene for a few days.

Feed wastewater characteristics

The synthetic wastewater employed in this study was formulated to simulate petrochemical industrial wastewater in terms of COD and styrene concentrations and had the following composition (Table1). This resulted in a wastewater which had a COD of 1500 mg/L and contained the elements required for proper growth of the mixed bacterial population in activated sludge.

Experimental setup

A schematic diagram of the MBR experimental rig used in the present study is presented in Fig.1. The bioreactor consisted of a 5L acrylic tank, in which a Kubota (Kubota corporation, Japan) flat sheet membrane was immersed. Specifications of the membrane is given in Table 2. The gap between the membrane and the wall was set at 7mm to obtain efficient scouring of the

Table 1: Synthetic wastewater composition

Parameter	Concentration(mg/L)	Parameter	Concentration(mg/L)
styrene	50	CoCl ₂	0.094
Ethanol	645	Na ₂ MoO ₄	0.423
K ₂ HPO ₄	32.9	MnSO ₄	0.014
KH ₂ PO ₄	42.14	FeCl ₃	5
NH ₄ Cl	286.6	ZnSO ₄	2
MgSO ₄ .7H ₂ O	13	EDTA	7
CaCl ₂ .2H ₂ O	7	NaHCO ₃	500
NiCl ₂	0.0094		

membrane by the air flow (Ndinisa *et al.*, 2006). Air was introduced at a distance of 10cm beneath the membrane module through a stainless steel tube containing ten 2mm nozzles. The output of level sensor, pressure transmitter, Dissolved Oxygen (DO)-meter, pH-meter, and thermometer were connected to a data acquisition system to enable monitoring of the process parameters via PC. Effluent turbidity was determined by using Hach 2100N Turbidimeter. The inlet and outlet peristaltic pumps were controlled (i.e. turned on or off) based on the data supplied by the level sensor. The synthetic wastewater was stored in a feed tank prior to being pumped to the MBR. In order to avoid the escape of styrene to the gas phase of the tank, the tank was separated into two parts using a 2cm thickness Teflon sheet; the bottom section was completely filled with

Table 2: Specifications of MBR system

Item	Specification
Membrane type	Flat sheet
Membrane material	Polyethelene
Membrane pore size	0.4μm
Membrane area	0.11m ²
Effective volume of reactor	5 L

the synthetic media, whereas a container full of styrene was placed in the upper part. Using a feed tank with this design ensured that the concentration of styrene in the inlet to MBR throughout its operation did not deviate from 50mg/L by more than 0.1%. This system operated at solid retention time of 25 day and HRT=24h and 18h and two SRTs were allowed to pass before data pertaining to the treatment performance was obtained. The DO concentration were 5.6 and 4.5 mg/L at the HRT of 24 and 18 h respectively.

Analytical methods

Styrene concentrations were analyzed by using a gas chromatograph (Younglin) equipped with a Helium Ionization Detector and a 50m long capillary column. The oven temperature was maintained at 70°C for 1min and raised to 140°C at 10°C/min. The temperatures of the injector and the detector were fixed at 200°C and 240°C, respectively. styrene concentration in the liquid phase was estimated using headspace method. Each gas and liquid sample was measured three times. The MLSS, MLVSS and COD were estimated according to Standard Methods (APHA, AWWA and WEF, 1998). SMP and EPS were measured using the method described by Chang *et al.* (1999) according to the following procedure: The mixed liquor of activated sludge was centrifuged in order to remove the bulk solution (3200 rpm, 30 min). The supernatant was separated and was named as "SMP". The remaining pellet was resuspended with 0.9% NaCl solution. This solution was heated at 100 °C for 1 h. The extracted solution was centrifuged again under the same operating condition as mentioned above. The supernatant obtained in this way was named as the "EPS solution". In this work, the sum of total proteins and polysaccharide was considered to represent the total amount of EPS and SMP because they are the dominant components typically found in extracted EPS and SMP. Protein fraction of SMP and EPS was measured using the folin phenol reagent method (Lowry *et al.*, 1951), whereas the corresponding polysaccharide fraction was determined by phenol-sulfuric acid method (Dubois *et al.*, 1956).

The sludge flocs were examined by light microscopy. The floc size distribution was determined by a Malvern Mastersizer 2000 instrument with a detection range of 0.02–2000μm.

RESULTS

COD and styrene removal performance of the MBR system

The results of the present study (Table 3) shows that MBR operated at SRT of 25 days and HRT in the range 18-24 hours gave over 99% removal of COD and styrene from a synthetic wastewater containing 50 ppm styrene with COD of 1500 mg/L. The concentration of styrene in the output

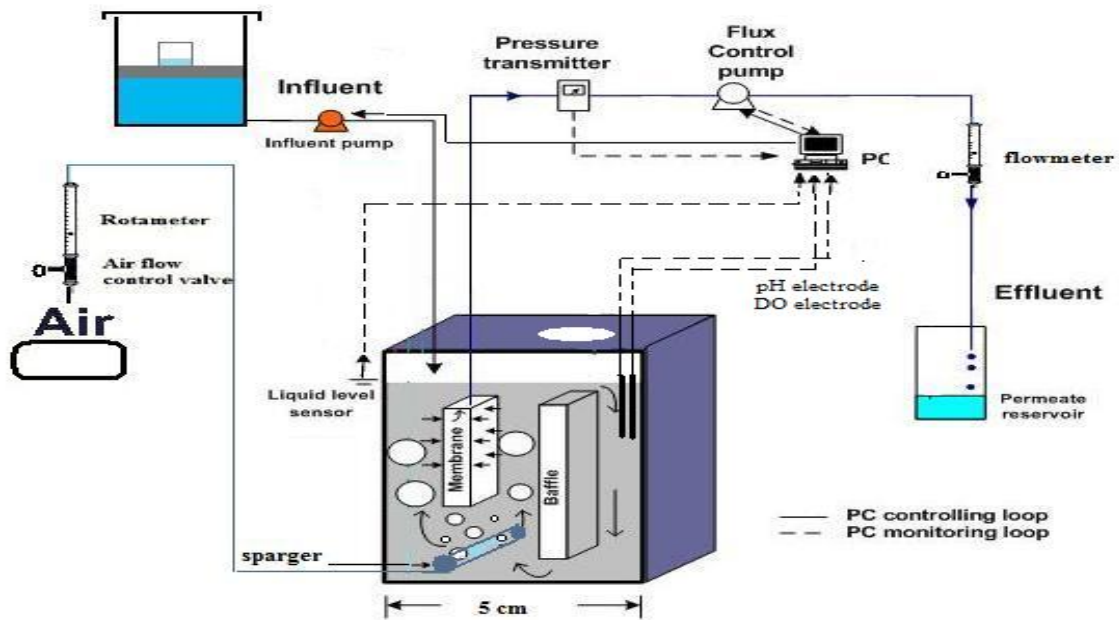


Fig.1: Schematic of the MBR

Table 3: The removal performance and volatilization of styrene and other VOCs and the COD removal performance obtained in the present work and previous studies in MBR and activated sludge processes

Process	Type of wastewater	Substrate	COD (mg/L)	HRT (h)	MLSS (mg/L)	COD removal (%)	Stripping (%)	Substrate removal (%)	Ref .
MBR	synthetic	styrene	1500	18-24	4300-5300	99	<1	99	This study
MBR	ABS	Acrilotiril	2950-4410	12-24	6-35000	90-98	2-10	-	Chang (2006a)
MBR	synthetic	vinylacetate - Acetaldehyde Butyraldehyde	7300	81.6-165.6	4400-11600	99.3	0.59-12.5		Min and Ergas (2006)
MBR	Petrochemical waste water	-	700-2000	13-19	8600-9600	92-95	-	-	Qin(2007)
CASP	synthetic	styrene	-	18-24	2040	-	15	85.7	Hsieh C.C, (2000).
CASP	Petrochemical waste water	Vinylchloride Ethylenechloride	200-850	-	-	59-89	-	45-92%	Shokrolahzadeh (2008)
CASP	publicly owned treatment works	Chlorinated and nonchlorinated VOCs*	115 (BOD)	5-6	1610-1770	87-97.2 (BOD)	2-83.1 16-22%	19.5-97%	Namkung and Rittmann (1987)
CASP	Mill creek wastewater with spike with VOC	VOC compound**	-	7.5	2000	69	1.1-100	0-93.4	Bhattacharya (1996)

* Benzene; Chlorobenzene; chloroform; 1,2-Dichloroethane; Ethyl benzene; Methylchloride; Tetrachloroethylene; Toluene;;1,2-Trans- Dichloroethane ;1,1,1trichloroethane ; trichloroethylene;

**Aceton; Mthyle ethyl keton; cyclohexanon; Tetrahydrofuran carbon; Tetrachloride; 1,1,1Trichloroethane; 1,1,2Trichloroethane Tetrachloroethylene trichloroethylene; Chlorobenzene; Ethyl benzene.

air from MBR was consistently below 0.1 ppm. Given the air rate used (7 L/min) the contribution of the stripping mechanism to overall styrene removal at HRT of 18 and 24 hours was 0.3% and 0.4% respectively. After steady state conditions were achieved, the MLSS at HRT of 18 and 24 hours was 5300 mg/L and 4300 mg/L respectively. The corresponding MLVSS values were 5100 mg/L and 4150 mg/L respectively. This gives a ratio of MLSS/MLVSS of 0.96 for both HRTs. Turbidity of the permeate at both HRTs was consistently below the detection limit of the Turbidity meter (< 1 NTU).

Fouling of membrane during operation of the MBR system

Fouling rate inside the MBR during its operation at both HRTs was quantified by monitoring the transmembrane pressure (TMP) values (results not shown). The range of TMP values, as well as the highest rate of rise of TMP with time during the operation of the MBR at HRTs of 18 and 24 hours, is presented in Table 4. The results show a significant increase in the membrane fouling rate when HRT is reduced from 24 to 18 hours.

Table4: Range of values of total EPS and SMP and TMP, as well as the highest rate of rise of TMP during the operation of MBR at two HRTs

HRT (h)	18	24
dTMP/dt(KPa/day)	0.12	0.012
Total EPS(mg/g MLVSS)	140-155	200-226
Total SMP(mg/L)	20-24	24-27

EPS and SMP

In Table 4, the range of total EPS and SMP values obtained during the operation of MBR at HRTs of 18 and 24 hours, is presented. It should be pointed out the values given are the range obtained during the operation of the MBR at the two HRTs and are not steady state values. The results show a significant decrease in the range of EPS values and a slight decrease in the range of SMP values, when HRT was reduced to 18 hours.

Particle size distribution

PSD data are presented in Fig. 2. These results show decrease in the mean particle size of the activated sludge when HRT was decreased to 18 hours.

DISCUSSION

COD and styrene removal performance of the MBR system

The results obtained in the present work shows a very good performance of the MBR in treating a styrene containing wastewater since very high overall COD and styrene removal with insignificant volatilization of the toxic styrene were obtained and the effluent had extremely low turbidity.

Comparison of the results obtained in the

present work with previous work in which either traditional activated sludge (Namkung and Rittman, 1987; Bhattacharya *et al.*, 1996; Heiseh, 2000; Shokrollahzadeh *et al.*, 2008) or MBR (Change *et al.*, 2006 a,b; Min and Ergas, 2006) processes have been used for treating with VOC containing wastewaters shows a much lower volatilization of a VOC (styrene in this study) compared to other studies (Table 3). For example, based on the concentrations of styrene measured in the outlet air after steady state conditions were achieved in the MBR, the contribution of stripping to overall styrene removal at both HRTs was $< 1\%$.

In the paper by Hsieh (2000) a model is presented for removal of VOCs in an activated sludge process. The model predicts that, at typical operating conditions of an activated sludge process, the contribution of the stripping mechanism to overall styrene removal from the wastewater is around 15%. Also, according to Hsieh (2000) adsorption to activated sludge does not significantly contribute to styrene removal. It can therefore be concluded that in the present study in the MBR, styrene was almost exclusively removed through biological degradation. The

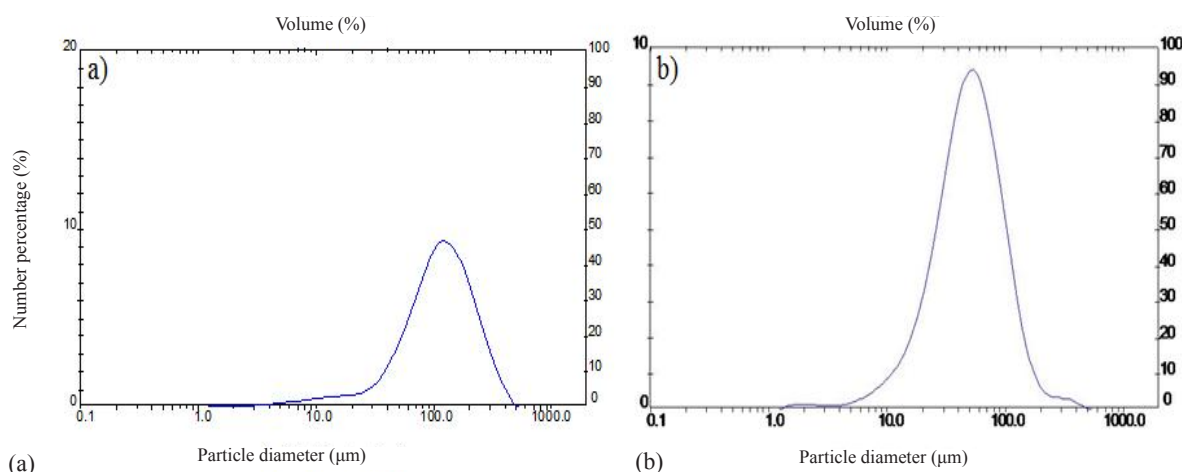


Fig. 2: PSD curves of activated sludge samples from the MBR operated at HRT of (a) 24 hours and (b) 18 hours

reason for the lower volatilization obtained in MBR compared to an activated sludge process can be understood by comparison of the MLSS data presented in Table 3. This shows that, due to higher SRT (25 days) employed in the MBR compared to that commonly achievable in traditional activated sludge processes, much higher MLSS were obtained in the MBR at both HRTs compared to the corresponding activated sludge process operated at the same HRTs.

The finding of other work in which MBR was used to treat other VOCs in an MBR is also presented in Table 3. These studies include the removal of acrylonitrile monomer from ABS industrial wastewater (Chang *et al.*, 2006 b) and a mixture of vinyl acetate-acetaldehyde-butanaldehyde from a synthetic wastewater (Min and Ergas, 2006). In both studies the contribution of stripping mechanism to VOC removal from the liquid phase was higher than that obtained in the present work, although still lower than the corresponding values that are obtained in a traditional activated sludge process. The better performance, in terms of reduced volatilization of a VOC, obtained in the present study relates to the lower solubility and/or lower volatility of styrene compared to the VOCs employed in the previous studies. However, the comparison of VOC removal data obtained in the present work or previously reported for MBR processes with those reported for an activated sludge process (Table 3) shows that the use of MBR for treating

VOC containing industrial wastewaters will result in lower volatilization compared to the treatment of the same wastewater in an activated sludge process. As reported in Table 3, the COD removal obtained in the MBR studies for both synthetic and real petrochemical wastewaters is also usually higher than those reported for activated sludge processes; this can again be attributed to the higher MLSS that can be achieved inside an MBR.

Another point concerns the toxic effect of styrene on biomass performance and properties. Toxic aromatic VOCs such as styrene are common constituents of petrochemical wastewaters. The measurements (results not presented) of specific oxygen uptake rates (SOUR) for the bacterial community in the seed activated sludge and activated sludge during the operation of the MBR at the highest OLR (corresponding to the lower HRT employed in the present study) showed no effect of styrene at this loading rate on the performance of bacterial community in activated sludge inside MBR. However, in previous studies toxic effects of styrene on bacterial activity has been reported. For example, Kuhlmeier (1989) has reported that styrene imposes significant bacterial inhibition at concentrations higher than 100 mg/L. Kinetic studies of styrene biodegradation in the liquid phase using both pure and mixed bacterial strains have shown that Haldane kinetics can be used to model the styrene biodegradation process in the liquid phase (Jung and Park, 2005; Jorio

et al., 2005). Haldane kinetics predicts that below a styrene concentration threshold, increase in styrene concentration increases the rate of styrene biodegradation, whereas at styrene concentrations above this threshold the opposite trend is observed (Shuler and Kargi, 2001). Kinetic studies with the activated sludge inoculums used in the present study also showed that, for the styrene biodegradation by the bacterial community inside this activated sludge, Haldane kinetics is a suitable model and the inhibitory styrene concentration is 21.57mg/L (Babae *et al.* 2009). Therefore, it is to be expected that at higher OLRs (i.e. lower HRTs or higher concentrations of styrene in the wastewater), styrene might result in the retardation of the activity of the bacterial population in activated sludge inside the MBR; this might result in a lowering of the performance of the MBR process for treating high OLR styrene containing wastewaters. This matter is presently under investigation in the authors' laboratory.

Fouling of membrane

The results obtained in the present work showed a significant increase in the membrane fouling rate when HRT is reduced from 24 to 18 hours (Table 3). The change in morphology from floccular to dispersed was confirmed both by microscopic observation of activated sludge samples from the MBR at the two HRTs, as well PSD curves for the two sludge samples (Fig. 2); the latter shows shift to smaller particle sizes when HRT was decreased from 24 to 18 hours. This change in morphology can be related to decrease in the EPS concentration (Table 4), since EPS is one of the factors that cause effective bioflocculation (Ng and Hermanowicz, 2005). This explanation is also confirmed by previous works in MBR (Ng and Hermanowicz, 2005; Massae *et al.*, 2006). Another factor contributing to the formation of dispersed morphology at the lower HRT could be increase in the styrene loading rate, since it has been previously reported that toxic factors can result in deflocculation of activated sludge (Li *et al.*, 1993; Bott and Love, 2001, 2002). Decrease in HRT from 24 to 18 hours resulted in significant increase in the rate of membrane fouling which was attributed to the deflocculation of the activated sludge. According to Bai and Leow

(2002) sludge particles having a size smaller than 50µm result in a greater specific cake resistance.

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