

ORIGINAL ARTICLE

Determining apparent first-order Biological Rate constant for Styrene and Ethylbenzene in MBR with Mathematical model

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ABSTRACT

In this paper, mathematical model was applied for determining styrene and ethylbenzene apparent first-order biological rate constant in a membrane bioreactor (MBR) process. To achieve this goal MBR with three different hydraulic retention time (HRT) of 10, 15 & 20 hr and two sludge retention time (SRT) of 20 & 10 day has been used. Then with comparison of removal rates that calculated through mathematical model and results that obtained from lab scale MBR, examined apparent first-order biological rate constant (K_b) for styrene and ethylbenzene. Results indicate that, K_b value for styrene and ethylbenzene is about 0.077 and 0.35, respectively. Model results were verified with comparison of three removal rates (stripping, biodegradation and adsorption to the sludge) that obtained from experimental data for the MBR process at various HRTs and SRTs. Also effect of SRT and HRT on styrene and ethylbenzene biological removal efficiency investigated. The experimental and mathematical model results show that for SRT of 20d, optimum HRT for biological removal efficiency is 15hr.

Keywords: *Mathematical model, Biological rate constant, MBR, HRT and SRT.*

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INTRODUCTION

Nowadays, one of the main issues facing the industrialized world is to minimize environmental contamination and to find out effective treatment methods. Over the last few decades, hazardous organic mixtures poured to the environment by human activities as well as industries. For almost 45 years, the issue of reducing emissions of volatile organic compounds (VOCs), such as styrene and ethylbenzene in the air completely comprised in a concept of sustainable progress, concerns the international community [1, 2]. Wastewater treatment methods have been developed to respond to the concerns about public health [3]. These methods include physical methods such as activated carbon adsorption [4], chemical methods as ozonation [5] and Biological methods such as conventional activated sludge process (CASPs), rotating biological contactor (RBC) processes [6, 7], and stabilized biofilm [8]. Biological methods compared to other methods have more advantages as they are more compatible to the nature and among the biological methods, conventional activated sludge is the only method that is not only used in petrochemical industry, but also it is widely used in many industries. It has been reported that the removal of volatile organic compounds in conventional activated sludge systems is done through three mechanisms: biological, absorption and stripping removal [2]. Due to low MLSS concentration in CASPs, they are one of the most significant VOC emission sources. Membrane bioreactor (MBR) is an activated sludge process in which the sedimentation unit is replaced with membrane. In the MBR process can be achieved to higher mixed liquor suspended solid (MLSS) concentration and higher apparent first-order biological rate constant (K_b); therefore in these systems reduce VOCs emission. The removal of VOCs in biological treatment wastewater systems is determined by three mechanisms including biodegradation, stripping, and adsorption to the waste sludge, which depend on chemical, physical properties of VOCs, such as Henry constant, its diffusion coefficient in water, and first-order biological rate constant and the

design/operational parameters, such as hydraulic retention time (HRT), sludge retention time (SRT), organic loading rate (OLR), dissolved oxygen (DO) concentration or influent air flow rate [9,10].

Defining the degradation kinetics of these contaminants by bacterial populations is one of the principle steps to forecast and to optimize the activated sludge processes at industrial scales. Mathematical models have been developed to evaluate the first-order biological rate constant styrene and ethylbenzene.

The main goal of this study is determining K_b for styrene and ethylbenzene in MBR. To achieve this aim a lab scale membrane bioreactor and the mathematical model has been used. Also the mathematical model was applied and relative stripping, biodegradation and adsorption rates were calculated at various HRTs and SRTs in a biological treatment process treating styrene and ethylbenzene. Further model results are verified by experimental data for the membrane bioreactor process at various HRTs and SRTs.

Modeling for estimating VOC Removal Efficiency

Hsieh [2] developed equations to determine VOCs removal rate in an activated sludge system using diffused aeration. The overall VOC removal is quantified by the ratio of the effluent VOC concentration to the influent VOC concentration. The total VOC removal rate can also be expressed with three removal fractions by stripping, biodegradation and adsorption mechanisms, as shown in Equation 1.

$$\text{Overall VOC removal efficiency} = 1 - \frac{C_e}{C_i} = \frac{R_s + R_{Bio} + R_{ads}}{1 + R_s + R_{Bio} + R_{ads}} \quad (1)$$

Where R_s , R_{Bio} and R_{ads} (dimensionless) are stripping, biodegradation and adsorption removal fractions, respectively; C_e (mg/L) is target compound effluent concentration; C_i (mg/L) is target compound influent concentration. Removal efficiency through stripping, biodegradation and adsorption mechanisms can be computed relative to each removal fraction. For example, the biodegradation removal fraction is calculated as shown in Equation 2.

$$\text{Removal efficiency through biodegradation mechanism} = \frac{R_{Bio}}{1 + R_s + R_{Bio} + R_{ads}} \quad (2)$$

Stripping removal fraction

The stripping removal fraction during aeration can be estimated by Equation 3. At constant oxygen transfer efficiency, the stripping removal fraction can be influenced by influent VOC concentration, the organic loading rate, and MLSS and dissolved oxygen (DO) concentration.

$$R_s = \frac{Q_g H_c S_d}{Q_L} \quad (3)$$

Where Q_g is air flow rate; H_c is Henry's law constant; S_d is VOC saturation in exit bubbles which is computed by Equation 4.

$$S_d = 1 - \exp\left(-\frac{K_L a_{VOC} V}{Q_g H_c}\right) \quad (4)$$

$$Q_g = \frac{1}{0.23 \rho_{air}} \left(Q_L (C_i - C_e) - 1.42 \frac{X V}{SRT} \right) \quad (5)$$

Where V is volume reactor and the $K_L a_{VOC}$ is VOC mass transfer coefficient that it can be estimated from overall oxygen transfer and the ratio of the diffusion coefficients as shown in Equations 6 and 7 [11].

$$K_L a_{VOC} = \left(\frac{D_{wVOC}}{D_{wO_2}}\right)^n K_L a_{O_2} \quad (6)$$

$$K_L a_{O_2} = \frac{(1 - Y) \left(\frac{S_i - S_e}{\theta}\right) + 1.42 K_d X}{C_{O_2}^* - C_{O_2}} \quad (7)$$

Where D_{wVOC} is liquid phase diffusion coefficient of VOC; D_{wO_2} is liquid-phase diffusion coefficient of oxygen; n is 0.5 for diffused aeration system; Y is cell yield coefficient (0.4 to 0.6 basis COD); S_i and S_e are influent and effluent COD concentration; X is MLVSS concentration; θ is hydraulic retention time; K_d is

endogenous biomass decay coefficient; $C_{O_2}^*$ and are DO saturation concentration that is about 9 mg/L; and C_{O_2} is DO concentration in the aeration basin.

Biodegradation removal fraction

The VOCs removal rate through biodegradation mechanism is independent of the VOC concentration at high VOC concentrations. For very dilute concentrations, the Monod kinetics rate becomes a first-order relationship with respect to VOC [2]. In this work, the first-order relationship was used to calculate biodegradation removal fraction as shown Equation 8.

$$R_{Bio} = \frac{K_b X V}{Q_L} = K_b \cdot X \cdot \theta \quad (8)$$

Where K_b is apparent first-order biological rate constant ($m^3/gVSS.d$) and X is active biomass concentration (MLVSS).

Adsorption removal fraction

The adsorption removal fraction to the waste sludge is dependent to the quantity of excess sludge as shown in Equation 9 [13].

$$R_{ads} = \frac{Q_w X_w K_p}{Q_L} = \frac{Q_w X K_p}{Q_L} = \frac{V X K_p}{SRT \cdot Q_L} \quad (9)$$

Where Q_w is excess sludge; and K_p is partition coefficient. The partition coefficient is proportional to the octanol-water partition coefficient (K_{ow}), as shown in Equation 10.

$$K_p = 6.3 \times 10^{-7} f_{oc} K_{ow} \quad (10)$$

Where f_{oc} is fraction of organic carbon in solids; and K_{ow} is octanol-water partition coefficient. The fraction of organic carbon in solids equals 0.53 for biological cells presented by $C_5H_7NO_2$.

MATERIALS AND METHODS

Experimental setup

The dimensions of the membrane bioreactor for this setup were of 60×22×6.5 cm. The effective volume in the reactor was 7L. The membrane used in this study was a Micro-Filtration (MF) type with an effective area of 0.1 m², pore nominal diameter of 0.4 μm, A4 sheet size. The membrane is produced by the Kubota® Company and is made of Poly-Ethylene (PE). The aeration process in MBR is done for two purposes, first is to supply the oxygen needed for biological processes, and second is to clean the membrane surface and reduce the fouling rate. To achieve the second goal, a poly (methyl methacrylate) (PMMA) plate was used as a baffle to keep the air bubbles near the membrane surface so that they can make proper tensions with it and wash the sediments out of the surface. The aerobic sludge used in the MBR basin was supplied from the activated sludge of the Tabriz Petrochemical Company then adapted with synthetic feed for one month.

Influent wastewater

The synthetic wastewater used in this research was formulated to simulate petrochemical industrial wastewater in terms of Chemical Oxygen Demand (COD), styrene, and ethylbenzene concentrations which are 1200, 100 and 100 mg/L respectively. Ethanol used as a carbon source which created a COD concentration of about 1200mg/L. The synthetic wastewater compositions used in the present study are described in Table 1.

Table 1. The synthetic wastewater compositions used in this work

components	Concentration (mg/L)
Ethanol	370
Styrene (STR)	100
Ethylbenzene (EB)	100
NH ₄ Cl	560
K ₂ HPO ₄	35
KH ₂ PO ₄	45
MgSO ₄ .7H ₂ O	13
CaCl ₂ .2H ₂ O	7
FeCl ₃	5
ZnSO ₄	2
NaHCO ₃	500
EDTA (C ₁₀ H ₁₆ N ₂ O ₈)	7

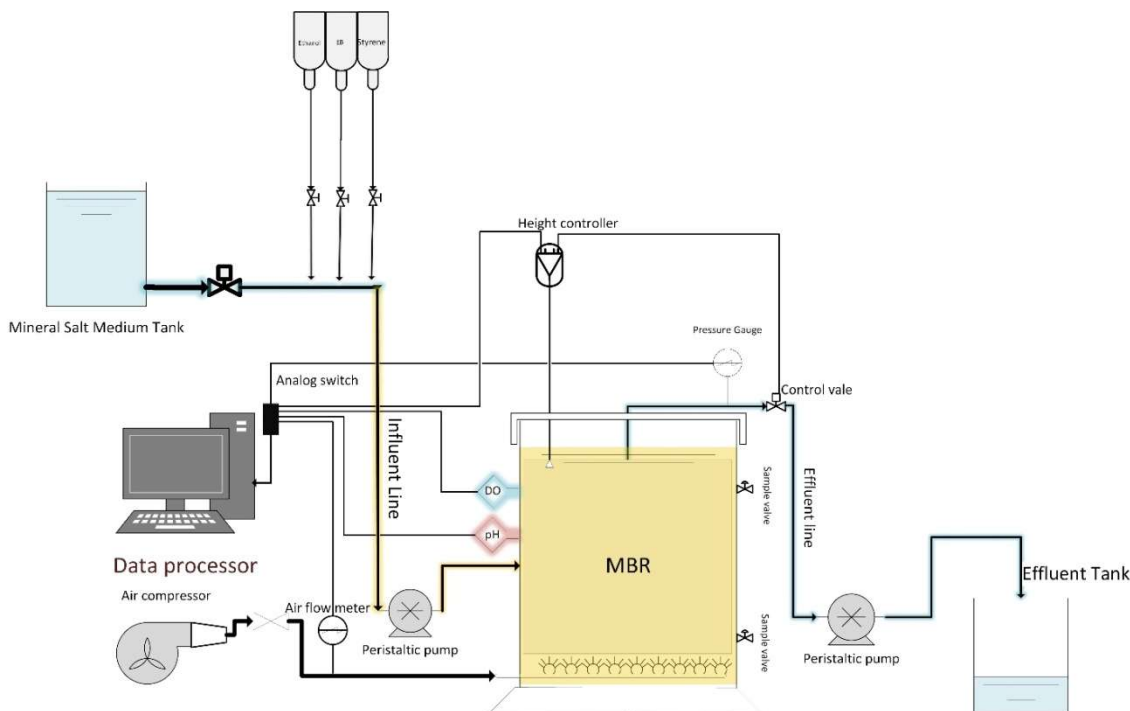


Fig. 1. Schematic of lab-scale experimental setup

Analytical methods and operation parameters

The styrene and ethylbenzene concentrations were analyzed using a Gas Chromatograph (GC). The GC (Young Lin, ACME-6100) was set with a Flame Ionizing Detector (FID) and the attached silica capillary column (DB-5, 0.53 mm I.D., 30 m length, 1 mm film thickness) that was designed to be well suited for the analysis of volatile components. The carrier gas was helium flowing at 15 mL/min. The oven temperature was maintained at 70 °C for 1min duration and raised to 140 °C. The temperatures of the injector and the detector were fixed at 200 and 240 °C, respectively. The Styrene and ethylbenzene concentrations in the liquid phase were estimated using the head-space method [14]. The gas flow rate from the bioreactors headspace was measured using a flow meter. The MLSS, MLVSS, and COD were estimated according to the standard methods [15].

The chemical and physical properties of styrene and ethylbenzene are presented in Table 2. Moreover, Table 3 provides the operating parameters for membrane bioreactor. An average 25°C temperature in the aeration basin was used throughout this study.

Table 2. The chemical, physical, properties of styrene and ethylbenzene at 25°C (Hsieh, 2000; MetCalf and Eddy, 2003)

properties	H (m ³ .atm/mol)	D _{w voc} (cm ² /s)	K _{ow}
Styrene	2.74 × 10 ⁻³	8.0 × 10 ⁻⁶	1444
Ethylbenzene	8.43 × 10 ⁻³	7.8 × 10 ⁻⁶	1349

Table 3. Operating parameters for membrane bioreactor

parameter	quantity	
	Model condition	Experimental condition
HRT	5 - 30 hr	10, 15 and 20 hr
SRT	5- 30 day	10 and 20day
COD	1200 mg/L	1200 mg/L
STR and EB concentrations	100 mg/L	100 mg/L
MLSS	2000-6000 mg/L	2000-6000 mg/L

Not : Styrene (STR), Ethylbenzene (EB)

RESULTS AND DISCUSSION

Experimental results for MBR

Styrene, ethylbenzene and COD removal efficiency in HRT=20 hr and SRT= 20 d

Styrene, ethylbenzene and COD removal efficiency is presented in Fig. 2. As can be seen, in steady state for HRT 20 hr, COD removal efficiency in reactor is around 98 percent and styrene, ethylbenzene removal is more than 99 percent. Moreover, the concentration of ethylbenzene and styrene in the reactor exit air was measured daily. The Fig.3. shows that in the hydraulic retention time of 20 hours the concentrations of ethylbenzene and styrene in reactor exit air, in steady-state is 0.7 ppm (equal 1.16% stripping removal) and 1 ppm (equal 1.65% stripping removal), respectively. This fact shows that the mechanism of removal in the reactor was not because of the volatility of styrene and ethylbenzene. Also, absorption of a pollution by a biomass, can only be considered as an important mechanism whenever partition coefficient of octanol - water ($\text{Log } K_{ow}$) is more than 4 [16], while this coefficient for styrene and ethylbenzene is about 3.15 and 2.85, respectively [2, 17]. Moreover, a study has shown that the absorption mechanism by sludge for styrene removal is too insignificant [13], therefore; the removal mechanism in the reactor is mainly through biological.

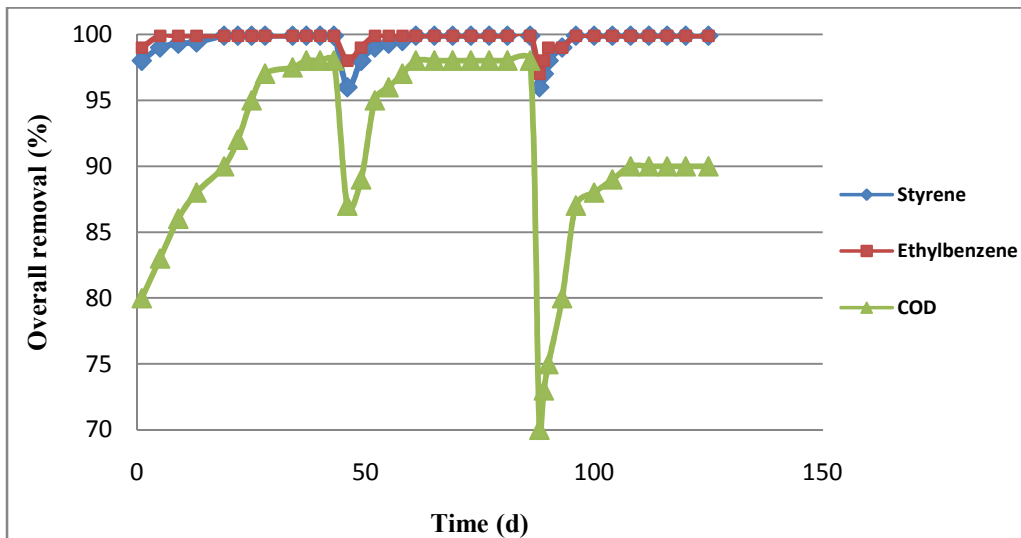


Fig. 2.Variations of COD, styrene and ethylbenzene removal during the operation of the MBR (HRT of 20h (days 0-43th), 15h (days 44-85th) and 10h (days 86-125th))

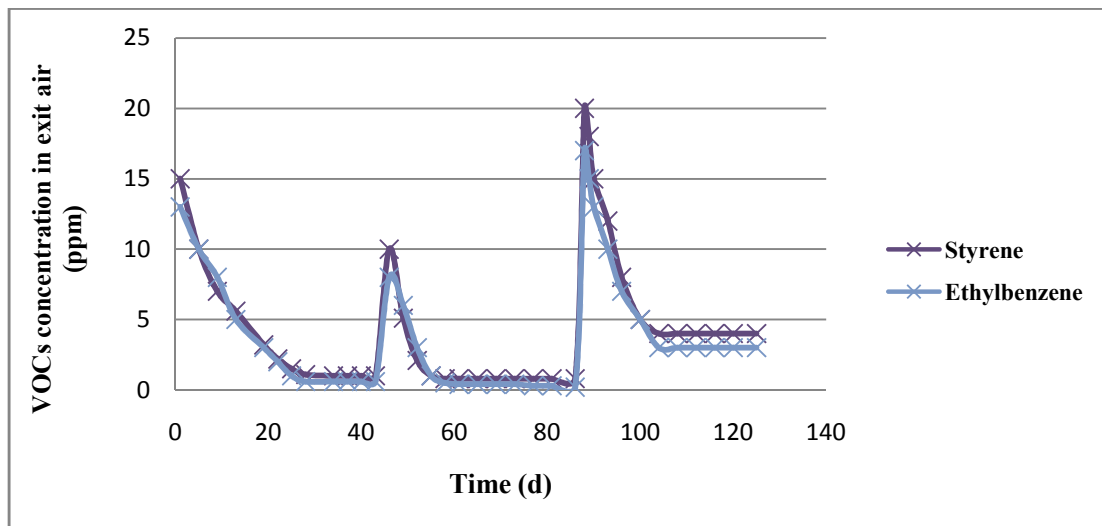


Fig. 3. VOCs concentration in the exit air during the operation of the MBR (HRT of 20h (days 0-43th), 15h (days 44-85th) and 10h (days 86-125th))

Styrene, ethylbenzene and COD removal efficiency in HRT=15 & 10 h and SRT =20 d

In the HRT of 15 hours, except during the first few days that the efficiency drops in MBR, the removal efficiency of COD, styrene and ethylbenzene increases and finally after reaching steady state respectively to 98, 99.9 and 99.9% increases. The reason of decrease in the beginning of day44th is for the increase of

organic load rate in the system, therefore; the microorganisms are under shock but after adapting themselves to the new condition, the efficiency of system gradually increases and eventually reaches to the steady state. As it is shown in Fig. 3, after the change of retention time of 20 to 15 hours, the stripping removal efficiency of styrene and ethylbenzene in the reactors light decreased, due to styrene and ethylbenzene concentrations slender decreased in the exit air from 1 and 0.7 ppm to 0.8 and .5 ppm, respectively. It has also been reported in previous studies that the removal through volatility reduced while HRT reduces (Chang et al, 2006).In fact, by reducing the retention time of 20 to 15 hours, biological removal efficiency of styrene and ethylbenzene has increased about 0.5 percent. Therefore; the result can be analyzed that when the retention time reduces, two parameters affect on the removal efficiency. First, organic loading rate increases and the amount of MLSS increases in rector. Second, the contact time of contaminants and sludge reduces. It is obvious that increasing of MLSS has a positive effect and reducing the contact time has a negative effect on the removal efficiency. But since MLSS concentration increased in the reactor, this leads to neutralize the negative effect of reducing the contact time and the removal efficiency increased.

Reducing the hydraulic retention time to 10 hours, according to the increased organic load rate in the system and on the other hand, the contact time between sludge and wastewater compared to the previous states (HRTs = 20 and 15h) became low, therefore biological removal efficiency in the reactor has reduced significantly. Although total removal efficiency of COD, styrene and ethylbenzene are 90, 99.9 and 99.9 percent, respectively, but biological removal for styrene and ethylbenzene are 93 and 94%. The cause of this decrease is due to a decrease in the concentration of MLSS in the system due to the increased organic loading rate. It should be noted that the concentration of exit gases also has increased. Styrene and ethylbenzene concentration in the exit air of reactor is 4 and 3 ppm, respectively, that is due to the decrease of MLSS concentration. Thus, in the retention time of 10 hours removal through biological process reduced than the two previous cases.

MLSS and MLVSS changes in the reactor (SRT= 20d)

In Fig 4. Changes in MLSS concentration versus time are shown. As it is clear in retention time of 20 hours, at the beginning the MLSS concentration increases and finally, at a concentration of 4,200 mg/L is fixed. As can be seen in Fig. 4, by reducing retention time and increasing of organic loading rate of 1.44 to 1.92 Kg / m3.d after a slight decrease in biomass concentration due to the shocked posed to system because of the increased loading rate, the biomass concentration starts to rise-up. Finally from the days 77th it reaches to its steady state. The MLSS concentration in the reactor is 4700 mg/L. Finally, when the retention time was reduced to 10 hours, not only the concentration of MLSS did not increase, but decreased to 2800 mg/lit. Changes of MLVSS are shown in Fig. 4, too. As can be seen, this parameter has almost the similar trend.

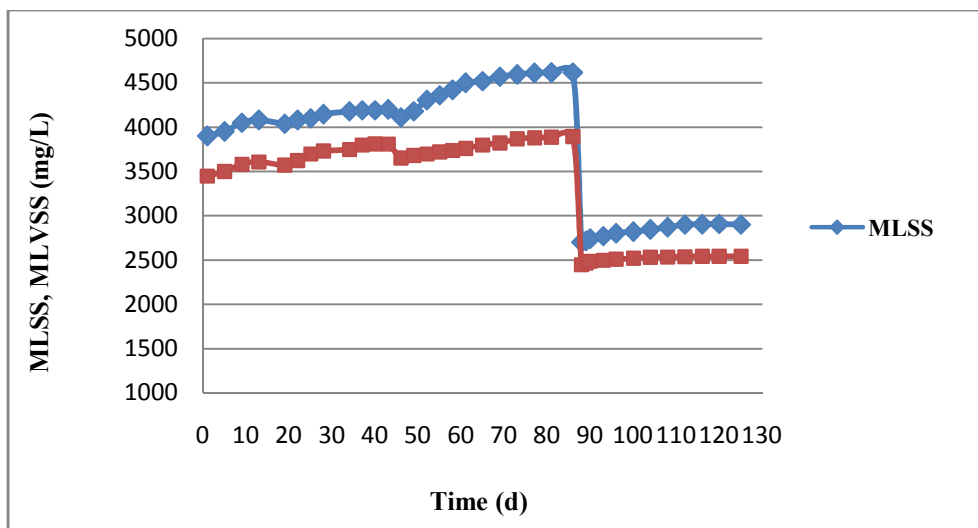


Fig. 4. Variations of MLSS and MLVSS during the operation of the MBR (HRT of 20h (days 0-43th), 15h (days 44-85th) and 10h (days 86-125th))

Styrene, ethylbenzene biological removal efficiency in HRT=15h and SRT = 10day

Styrene and ethylbenzene biological removal efficiency at SRT of 10 and HRT of 15h was measured in steady state. Styrene and ethylbenzene biological removal was about 94.6 and 98.7 percent; while for SRT

of 20d, was about 99 and 99 percent, respectively. Moreover, the concentration of ethylbenzene and styrene in the reactor exit air was measured daily in this condition. The results show that in SRT of 10 day the concentrations of ethylbenzene and styrene in reactor exit air, are 3.2 and 4.9 ppm, respectively. Compression of biological removal efficiency and VOCs concentration for styrene and ethylbenzene in two SRTs (10 & 20) presented in Table 4.

Table 4. Compression of biological removal efficiency and VOCs concentration in the exit air for two SRTs

SRT	Biological removal of STR	Biological removal of EB	STR conc. in air	EB conc. in air
10d	93.6	95.7	4.9	3.2
20d	98.5	99.1	0.8	0.5

Not : Styrene (STR), Ethylbenzene (EB)

Model results

Determine of K_b for styrene and ethylbenzene

According to the model developed in section 2, the percentage contribution of stripping, biodegradation and adsorption mechanisms calculated for styrene and ethylbenzene removal at varying apparent first-order biological rate constant at fixed HRT of 15hr and SRT of 10d (See Fig. 5 for styrene and Fig. 6 for ethylbenzene). The value of apparent first-order biological rate constant (K_b) for styrene and ethylbenzene was estimated by comparison of removal rates with experimental results in this condition. The K_b value for styrene and ethylbenzene examined about 0.077 and 0.35, respectively. Reference value for these materials is 0.08 and 0.4 respectively [18, 12].

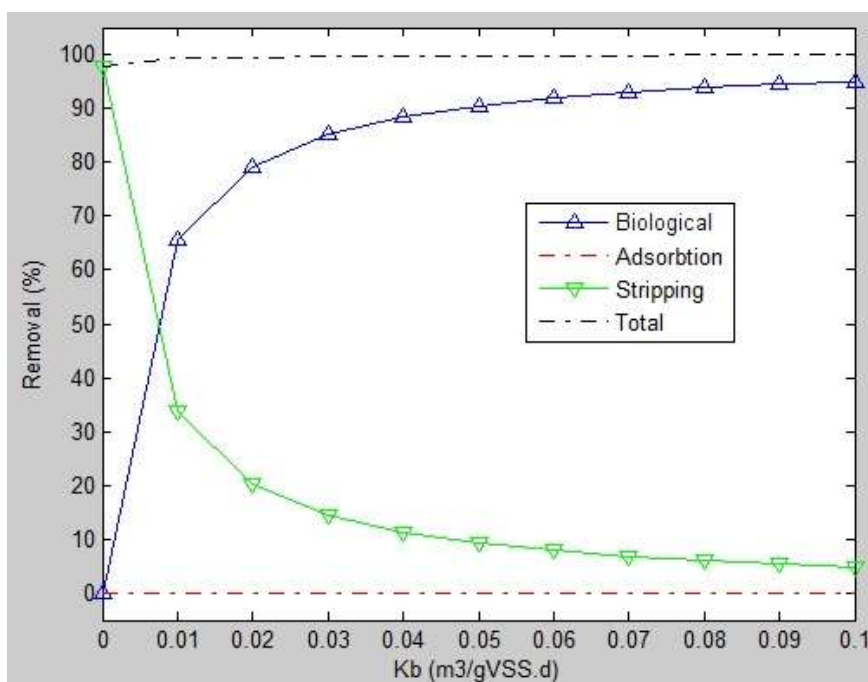


Fig. 5. Stripping, adsorption and biological removal efficiency for styrene in varying K_b at HRT =15h and SRT= 10d)

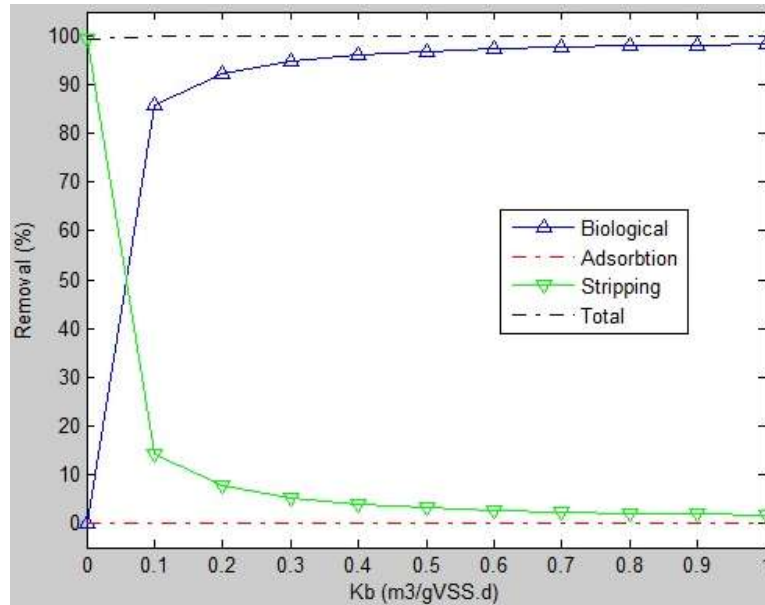


Fig. 6. Stripping, adsorption and biological removal efficiency for ethylbenzene in varying K_b at HRT = 15h and SRT = 10d

Effect of SRT and HRT in removal mechanism

Individual stripping, biodegradation and adsorption removal fractions were estimated using the equations presented in sections 2. The HRT and SRT values were varied. Overall and individual removal rates were then calculated using Equations 1 and 2. Since styrene has the strongest tendency to volatilize, the estimation results with styrene are presented in this section. Results for the ethylbenzene were presented along with the lab-scale MBR experimental results for comparison. Throughout all the calculations, it was assumed that influent and effluent VOC concentrations could be represented as COD.

Fig. 7. shows the calculated removal rate (biological, stripping and adsorption removal rate) at varying SRT and a fixed HRT of 15h for styrene. The overall removal rate is more than 99.9% at SRT of 5 to 30 days. The oxygen requirement and air flow-rate did not change at varying SRT. However, when SRT increases from 5 to 30d, increasing of MLSS concentration, leads to an increase in biodegradation rate from 89.2 to 94.1%, and a decrease in volatilization rate from 11.2 to 5.13%.

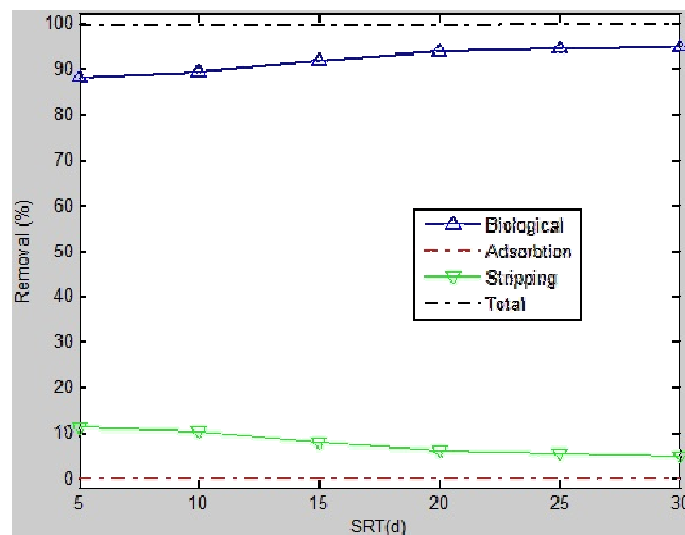


Fig. 7. Stripping, adsorption and biological removal efficiency for styrene in varying SRT and HRT=15h

HRT is one of the important parameters of biological processes such as MBR in petrochemical wastewater treatment [15, 19]. The biological treatment of this type of wastewater requires long HRT. For example, a CASP in a typical ABS manufacturing plant in the south Taiwan was designed with a HRT of

approximately 4 days; but this parameter was reported about 13-24hr for MBR processes (Chang et al., 2006).The Fig. 8.presents the distribution of three removal mechanisms for styrene at varying HRT at fixed SRT of 10 and 20 days. Similarly, the overall removal rate was greater than 99.9% except in HRT of 5-7 hr. The low HRTs (5-10hr) lead to a decrease in the MLSS concentration and increase in the DO concentration. Therefore removal efficiency through biodegradation mechanism in these HRTs is lower than HRTs of 15 to 30 hr. When HRT increases from 15 to 30hr for SRT of 20d, due to decreasing OLR the biodegradation rates decreased from 95.6 to 93.3% and the stripping rate increased from 4.9 to 6.3%. But for SRT of 10 d biodegradation rate decrease in this zone (HRT of 20 to 30hr).

The mathematical model can be a starting point to determine the optimum HRT to minimize VOC emissions from a biological treatment process. Optimum HRT for styrene and ethylbenzene biological removal is 20hr at SRT of 10day and 15 hr at SRT of 20 day.

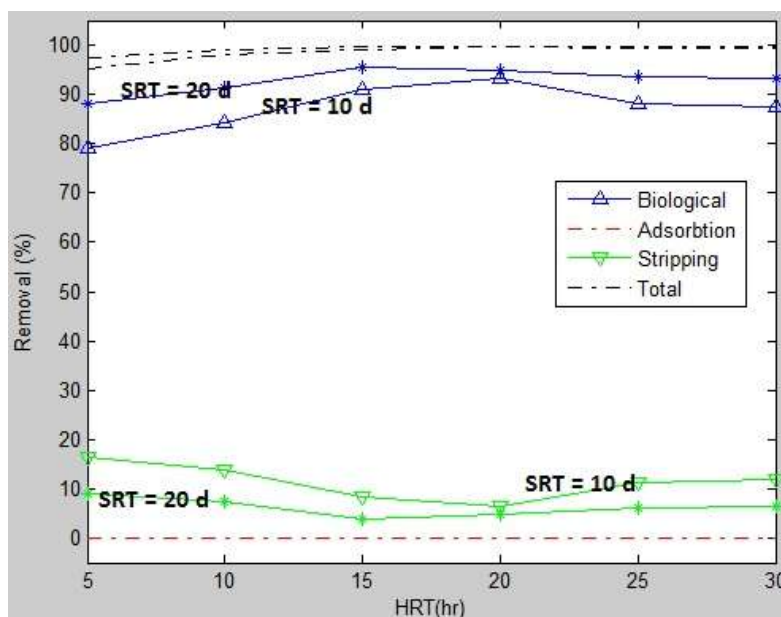


Fig. 8. Stripping, adsorption and biological removal efficiency for styrene in varying SRT and HRT

Comparison of experimental and model results

Table 5 summarizes the VOCs removal efficiency at varying HRTs and fixed SRT of 20d in both the lab-scale MBR and the mathematical modeling results. According to the experimental results, the overall VOC removal rate was more than 99.5% at three HRTs. Decreasing HRT from 20 to 15 hr led to an increase in MLSS concentration from 4100 to 46000 mg/L. But with decreasing HRT from 15 to 10hr, OLR significantly increase and MLSS concentration decrease to 2800mg/L, therefore biological removal efficiency drop. In the case of ethylbenzene, the experimental results showed that the biological removal efficiency increased from 98.3 to 99.1% and the stripping removal efficiency decreased from 1.2 to 0.8% by decreasing HRT from 20 to 15hr. Regarding styrene biodegradation removal efficiency in the lab-scale MBR had a low decline compared to ethylbenzene at three HRTs, as expected based on the modeling results. Table 6 Also present the VOC removal efficiency at varying SRTs and fixed HRT (HRT=15hr) in both the lab scale MBR and the mathematical modeling results. The both results (experimental and model) show that with decreasing SRT, biological removal rate for two VOCs was decreased.

Table 5. The VOC removal efficiency at varying HRTs and SRT=20d in both the lab-scale MBR and the mathematical model results

Removal (%)	Biological			Stripping			
	HRT(h)	20	15	10	20	15	10
Compounds							
STR (Exp)		98.2	98.9	93.3	1.7	1.3	6.6
STR (Model)		95.6	94.7	91.3	5.1	4.3	7.5
EB (Exp)		98.3	99.1	94.9	1.2	0.8	5.0
EB (model)		97.6	97.9	91.3	2.2	1.8	8.4

Not : Styrene (STR), Ethylbenzene (EB)

Table 6. The VOC removal efficiency at varying HRTs and SRT=20d in both the lab scale MBR and the mathematical model results

Removal (%)	Biological		Stripping	
	20 d	10 d	20 d	10 d
SRT				
Compounds				
STR (Exp)	98.5	93.6	1.4	6.3
STR (Model)	95.6	90.9	3.9	8.3
EB (Exp)	99.1	95.7	0.8	4.2
EB (model)	96.7	91.2	2.8	8.3

Note: Styrene (STR), Ethylbenzene (EB)

CONCLUSION

The results of mathematical model and experimental data revealed that can easily examine the value of apparent first-order biological rate constant for VOCs. The operation of a lab scale MBR confirms that a MBR can be a probable procedure to reduce VOC emissions from petrochemical wastewater. The experimental results show that for SRT of 20d, optimum HRT for biological removal efficiency is 15hr. The model results proved that optimum HRT for styrene and ethylbenzene biological removal is 15 hr at SRT of 20 day. Model results also show that optimum HRT is 20hr for SRT of 10 day.

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