ROLE OF MOVING BED BIOFILM REACTOR AND SEQUENCING BATCH REACTOR IN BIOLOGICAL DEGRADATION OF FORMALDEHYDE WASTEWATER

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ABSTRACT
Nowadays formaldehyde is used as raw material in many industries. It has also disinfection applications in some public places. Due to its toxicity for microorganisms, chemical or anaerobic biological methods are applied for treating wastewater containing formaldehyde.In this research, formaldehyde removal efficiencies of aerobic biological treatment systems including moving bed biofilm (MMBR) and sequencing batch reactors (SBR) were investigated. During all experiments, the efficiency of SBR was more than MBBR, but the difference was not significant statistically. According to the results, the best efficiencies were obtained for influent formaldehyde COD of 200 mg/L in MBBR and SBR which were 93% and 99.4%, respectively. The systems were also capable to treat higher formaldehyde concentrations (up to 2500 mg/L) with lower removal efficiency. The reaction kinetics followed the Stover-Kincannon second order model. The gram-positive and gram-negative bacillus and coccus as well as the gram-positive binary bacillus were found to be the most dominant species. The results of ^{13}C-NMR analysis have shown that formaldehyde and urea were converted into *N*-{[(aminocarbonyl) amino] methyl}urea and the residual formaldehyde was polymerized at room temperature.

Key words: Aerobic treatment; Formaldehyde; Moving bed reactor; Sequencing batch reactor; Kinetic coefficients; ^{13}C-NMR

INTRODUCTION
Formaldehyde has many industrial and non-industrial applications. It is used in production of resins, adhesives and hardboards, fungicides, pharmaceutics, paper, etc (Chen *et al*, 2010; Kumar *et. al*, 2007; Tejado *et al.*, 2007; Baraka *et al.*, 2007). This compound (with $KMnO_4$) is applied to sterile sanitary and medical parts (WHO, 1989).

Formaldehyde in low concentrations (less than 20 mg/L or 2%) causes incitement of mucous membrane, cough and disorder in gulping. At the same concentrations and longer exposure times, it increases danger of asthma. Inhalation of formaldehyde with concentration more than 0.2 mg/d may lead to extensive pain, wound, squirt, blood vomiting, vertigo and insufficiency in the blood system. Direct contact at formaldehyde concentration over 2% causes skin sensitivity. It has also been reported that exposure to formaldehyde can increase risk of cancer, especially lung, noise and blood ones (WHO, 1989). Therefore, any wastewater containing formaldehyde is a serious problem that should be treated before discharging to the environment.

Chemical and anaerobic biological methods are often applied for treating wastewaters containing formaldehyde because of its toxicity for microorganisms. Recently, some methods such

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as Fenton and photo-Fenton (Liang *et al.*, 2010; Kajitvichyanukul *et al*, 2006), photocatalytic degradation (Ge *et al.*, 2006), combination of chemical-biological (Lotfy *et al.*, 2002) and anaerobic-aerobic biological (Motelab *et al.*, 2002; Pereira *et al.*, 2008) have been reported.

In researches based on chemical treatments, it has been shown that each method of UV/H_2O_2, Fenton, photo-Fenton and their combination can completely remove formaldehyde solution with concentration of 1/3 M during 80 minutes retention time (Kajitvichyanukul *et. al*, 2006). Degradation of 50.3% formaldehyde with concentration of 10 µg/L has also been obtained by a thin layer of Ag/$InVO_4$-TiO_2 (Ge *et al.*, 2006). Advanced bioreactors like AFBGAC (Motelab *et al.*, 2002), ASBBR (Pereira *et al.*, 2008), combination of AUSBR and AASR (Eiroa *et. al*, 2006), combination of BAF and wetland (Melian *et al.*, 2008), SMBR (Fallah *et al.*, 2010, Naghizadeh *et al.*, 2008) and SBR (Mahvi, 2008), RO and NF (Zazouli *et al.*, 2008) were used for treating different municipal and industrial wastewaters. For example, From 97.4 to 99.9 and 99 percent formaldehyde removal efficiencies have been reported using AFBGAC and ASBBR, respectively (Motelab *et al.*, 2002, Pereira *et al.*, 2008). Over 99.7% removal efficiencies of formaldehyde and formic acid with concentrations of 2087 to 2200 and 1423 to 1599 mg/L using AUSBR combined with AASR have been achieved (Eiroa *et al.*, 2006). It has also been found that combination of BAF and wetland has removed 100% of formaldehyde with concentration of 200 to 817 mg/L (Melian *et al.*, 2008). Modern modifications of conventional activated sludge such as SBR (Mahvi, 2008), USBF (Mahvi et. al, 2008), MB-SBR (Hosseini Koupaie et. al, 2011) have been used for treating different kinds of wastewater.

In the late 1900s, Moving Bed Biofilm Reactor (MBBR) was introduced for biological treatment of different types of wastewater. Recently, it has been successfully used in treating different domestic and industrial wastewaters (Borghei *et al.*, 2004; Labelle *et al.*, 2005; Rusten *et al.*, 2006; Ayati *et al.*, 2007; Plattes *et al.*, 2007; Delnavaz *et al.*, 2008). It is a suitable alternative for common activated sludge reactors in treating domestic and industrial wastewaters in commercial scale. Currently, there are more than 400 units of full scaled wastewater treatment plants based on this process (Delnavaz *et al.*, 2008).

For the first time charged and discharged reactors have been introduced by Ardern and Locket (1914) which was later called as Sequencing Batch Reactor. These reactors were substituted by activated sludge process, because of requiring many laborers and diffusers frequent obstruction. In 1950, the first significant renewal of these systems was accomplished by Hoover and his colleagues in America (Irvine and Davis, 1971). Up to now, SBR has been successfully used for removing biological oxygen demand (BOD), chemical oxygen demand (COD), nitrogen, phosphorus (Mahvi *et al.*, 2005; Mahvi *et al.*, 2004), cyanide (White and Schnabel, 1998), nickel and plumb (Mahvi, 2008; Sirianuntapiboon and Ungkaprasatcha, 2007).

Most of the investigators have shown the application of chemical and or anaerobic processes in treating formaldehyde wastewater. Only in few researches, aerobic biological processes have been used to study the effect of low formaldehyde concentration on the removal rate of COD and total kjehldahl nitrogen (TKN) (Eiroa *et al.*, 2004; Garrido *et al.*, 2001; Campos *et al.*, 2003; Eiroa *et al.*, 2006). Therefore, the possibility of formaldehyde removal using MBBR and SBR aerobic treatment systems were the major purpose of this study.

MATERIALS AND METHODS

One MBBR and two serial SBRs were used in this research; each reactor had effective volume of 5 liters (Fig. 1). The characteristics of the bioreactors are given in Table 1.

Table1: Physical characteristics of the bioreactors

Parameter	Amount
Material	plexi glass
wall thickness (mm)	4
internal diameter (cm)	10
height (cm)	70
total volume of each pilot (L)	5.5
effective volume of each pilot (L)	5

Fig. 1: Pilots used in the research

Both systems were operated in batch-flow mode with 48 hours retention time. For SBR system, retention time for each reactor was 24 hours as 60% of the treated wastewater in the first reactor entered into the second one after 24 hours. In operation of both systems, filling and decanting steps were done manually.

In biofilm processes, the media has a great influence on treatment efficiency. The characteristics of bee cell 2000 media used in MBBR are given in Table 2.

Table 2: Media characteristics

Parameter	Amount
Material	HDPE[11]
Density (g/cm^3)	0.96
Effective surface of a medium (mm^2/piece)	857
specific surface (m^2/m^3)	1530
Total surface of a medium (mm^2/piece)	1800
Number (# / L)	361
mean weight (kg/m^3)	140

High Density Poly Ethylene

After adaptation step, the organic loading rate (OLR) was increased gradually. Influent feeding to the reactors during the study for both MBBR and SBR systems are presented in Figs. 2 and 3, respectively. In both Fig.s, parts "A", "B", and "C" indicate the influent feeding during pre-adaptation, adaptation and main experiments periods, respectively. Formaldehyde loading rate and related organic loading rate are given in Table 3.

Table 3: Formaldehyde and organic loading rates relation

OLR (g COD/L.d)	Formaldehyde loading (g Formaldehyde/L.d)
0	0
0.005	0.005
0.01	0.009
0.02	0.018
0.03	0.028
0.04	0.037
0.05	0.046
0.06	0.056
0.07	0.065
0.08	0.075
0.09	0.084
0.1	0.094
0.15	0.141
0.225	0.211
0.3	0.281
0.45	0.422
0.65	0.609
0.75	0.703
0.9	0.844
1	0.936
1.15	1.078
1.25	1.172
1.5	1.406

During the adaptation period, a combination of formaldehyde using formalin solution (37%) and glucose was used as feed for the reactors. During this period, 10% increase of formaldehyde followed by the same decrease in glucose amount was done in each step of system feeding. In the main experiments, only formaldehyde was used as the feed for the reactors. Urea, K_2HPO_4 and KH_2PO_4 were used as nitrogen and phosphorus sources, respectively. Mixture of $MgSO_4.7H_2O$ (5 mg/L), $CaCl_2$ (3.75 mg/L), $FeCl_3.6H2O$ (1 mg/L) and $Na_2MoO_4.2H_2O$ (1.26 mg/L) was also used as micro-nutrients.

All of the experiments were accomplished in accordance with the instructions presented in the "Standard Methods for the Examination of Water and Wastewater" (APHA, 2005). After investigating all methods, the best one was selected regarding to the laboratory facilities. For example, removal efficiency was determined using both formaldehyde concentration (using iodometric method (Lotfy *et. al*, 2002)) and COD variation ways. According to the results, removal of 0.94 mg/L formaldehyde caused decreasing of 1 mg/L COD. Therefore, variation of COD or formaldehyde concentration can be considered as criteria of removal efficiency. In this study, COD results were reported.

Controlled and measured parameters in the research were as follows:

Daily measured parameters:

- pH between 6.5 to7.5 using phosphoric acid or sodium hydroxide
- Dissolved oxygen between 3 to 3.5 mg/L
- Soluble COD to determine the removal efficiency

Periodically measured parameters:

- BOD_5 to investigate degradiablity of the pollutant
- MLSS (Mixed liquor suspended solid) and MLVSS (Mixed liquor volatile suspended solid) to investigate the growth ratio of attached and suspended microorganisms. The attached biomass was first desquamated by washing with water, and then mixed liquid was filtered through 0.45 µm millipore filter and dried at 105 ^{o}C for measuring dry weight (Chen *et al.*, 2008).

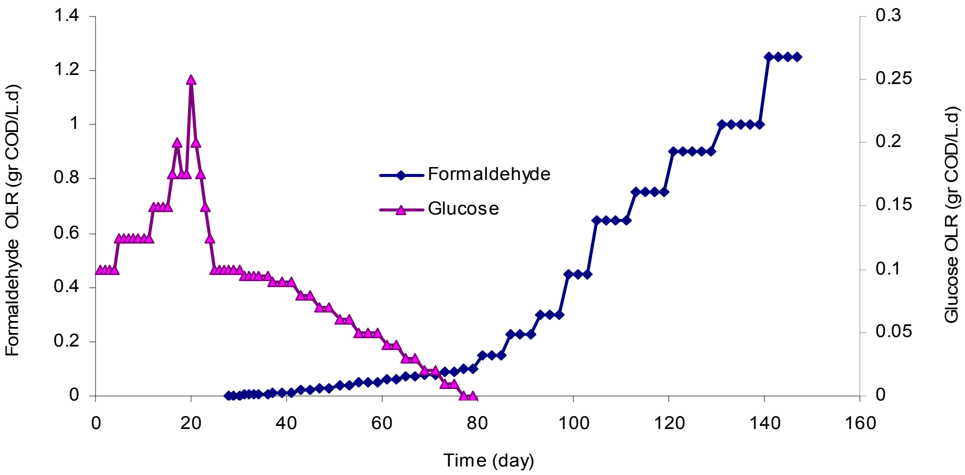

Fig. 2: Organic loading rate in MBBR

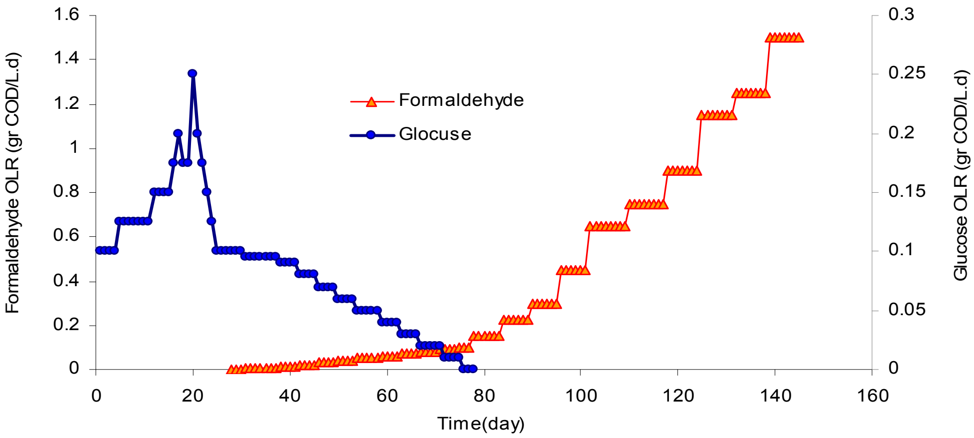

Fig. 3: Organic loading rate in SBR

- SVI (Sludge volume index) to evaluate sludge sedimentation properties
- Microbial culture, gram staining and microscope investigation to determine the type of microorganisms.

Finally, the effect of organic shock on the efficiency of each reactor was studied. Biodegradation of influent wastewater was also checked using carbon nuclear magnetic resonance (CNMR) spectrum analysis and the results were compared with those of ACD Labs program (2.70/01 July 1997).

RESULTS

Removal efficiency in the adaptation period

In this study, due to anti-microbial characteristics of formaldehyde, the adaptation period took about two months. Within this period, there weren't any media in MBBR system and it was applied as a SBR. In Fig. 4, the removal efficiencies of both systems during adaptation period are compared.

Removal efficiency in the main experiment

The efficiencies of MBBR and SBR systems in different influent CODs and retention times

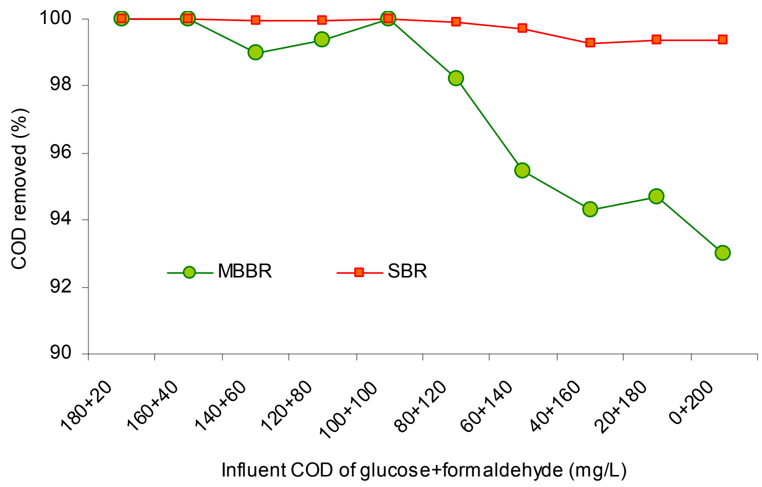

Fig. 4: MBBR and SBR efficiencies during the adaptation period

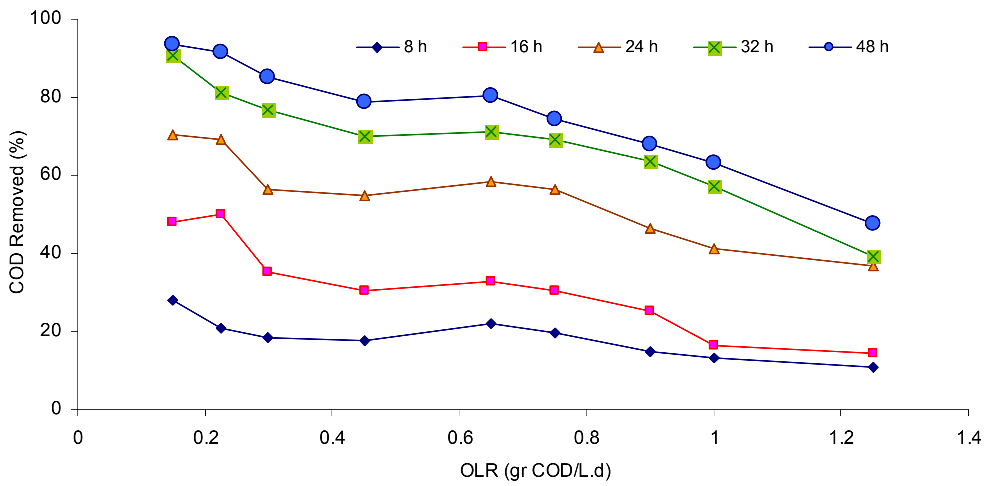

Fig. 5: MBBR efficiency in the different retention times

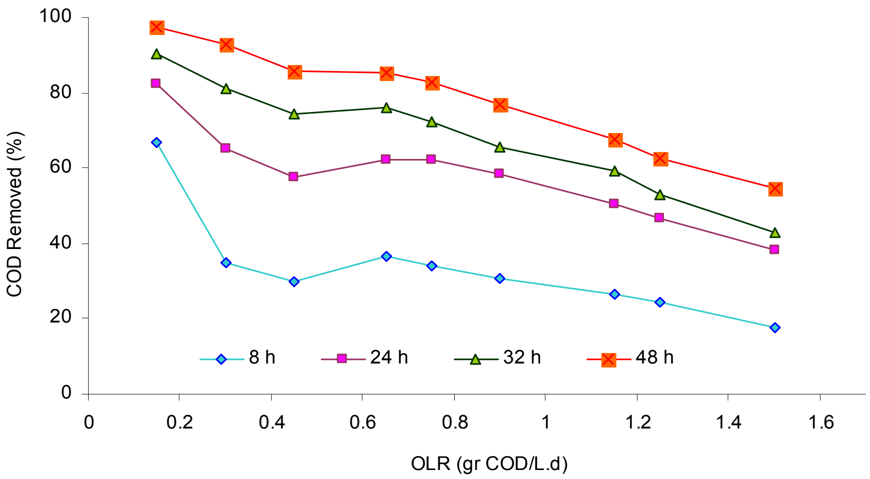

Fig. 6: SBR efficiency in the different retention times

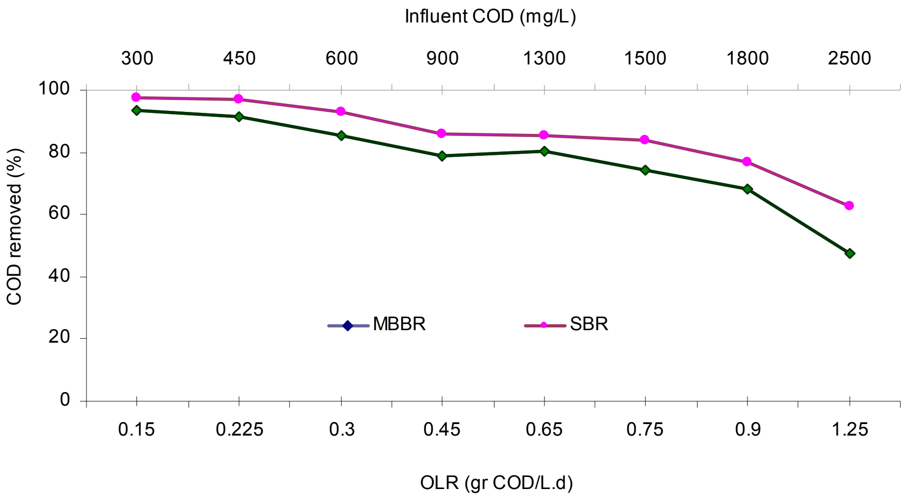

Fig. 7: Comparison of MBBR and SBR in the retention time of 48 hours

are presented in Figs. 5 and 6, respectively. Comparison of MBBR and SBR in the retention time of 48 hours is also shown in Fig. 7.

Carriers filling ratio effect in MBBR

The effect of carriers filling ratio on MBBR removal efficiency are shown in Fig. 8.

Shock loading effect on systems removal efficiencies

Figs. 9 and 10 show removal efficiency of both systems after organic shock loading.

Evaluation of control parameters

According to the parameters measured during the operation, MLVSS to MLSS ratio was equal to 0.75±0.04 and BOD_5 to COD ratio for MBBR and SBR were in the range of 0.54 to 0.75 and 0.42 to 0.77, respectively.

CNMR spectrum analysis

The results of CNMR spectrum analysis for the influent and effluent samples of the reactors are given in Figs. 11 and 12, respectively.

Kinetic investigation of biological reaction

According to the kinetic investigation of the reactions, no acceptable determination coefficient was achieved for the first order model and between Grau and Stover-Kincanon as the second order models, only the latter one was acceptable for all reactors.

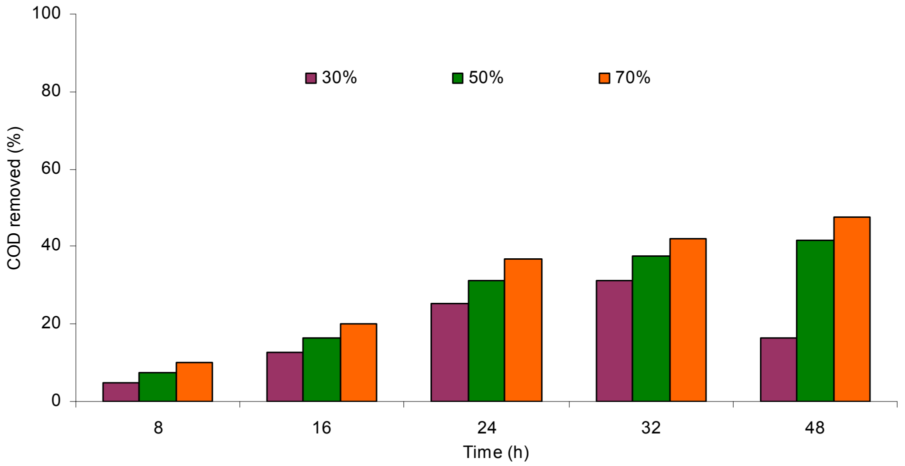

Fig. 8: Carriers filling ratio effect on MBBR

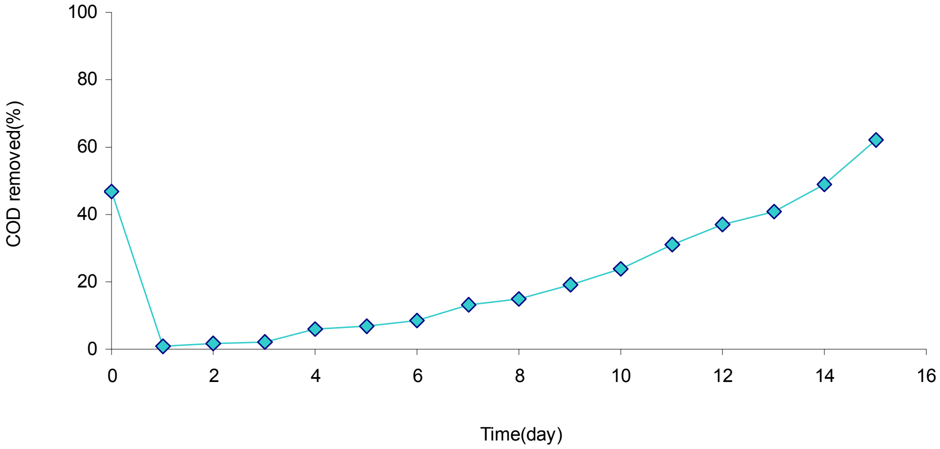

Fig. 9: Removal efficiency of MBBR after organic shock loading

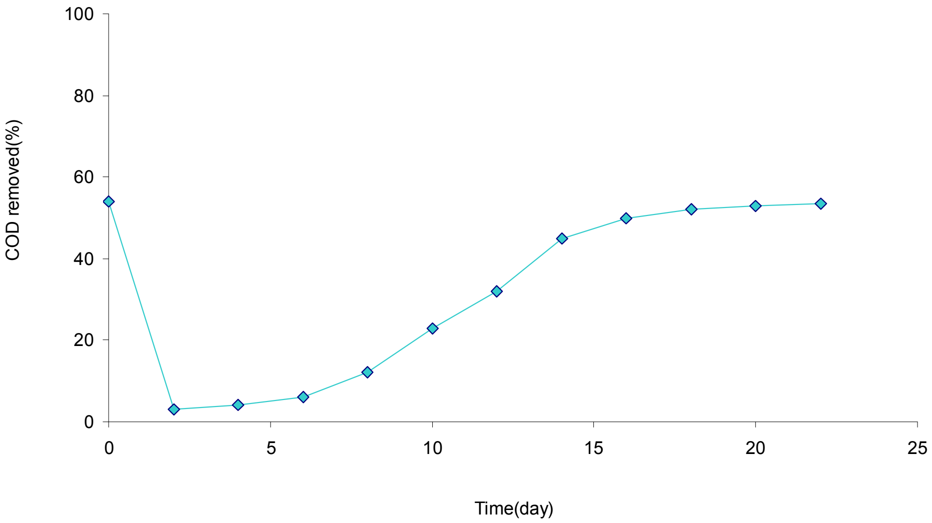

Fig. 10: Removal efficiency of SBR after organic shock loading

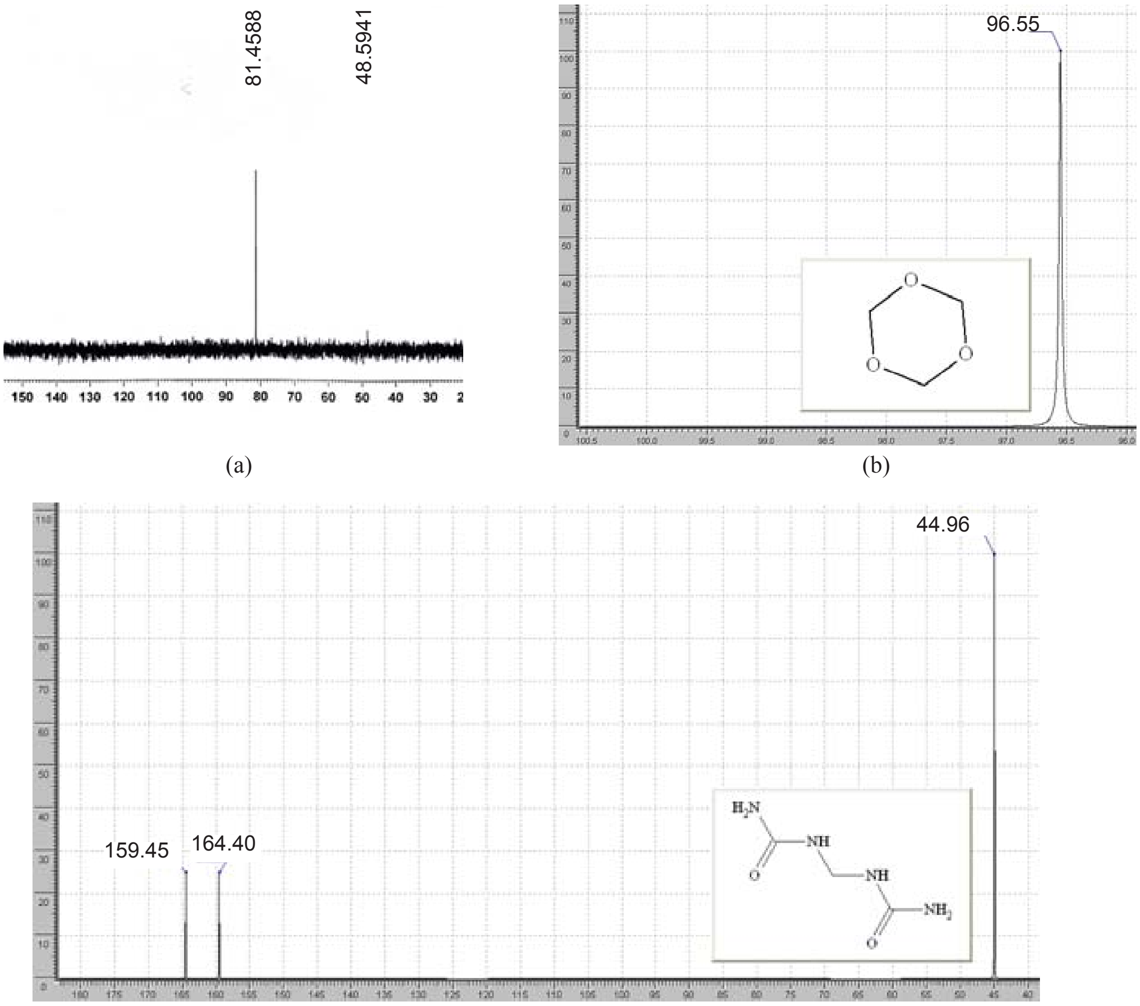

Fig. 11: CNMR analysis for the influent sample a)CNMR spectroscopy result for the influent sample;b) Analysis of formaldehyde polymer (1, 3, 5-trioxane) by ACD Labs program; c) Analysis of N-{[(amino¬¬¬ carbonyl) amino] methyl} urea by ACD Labs program

As shown in Figs. 13 to 18, the determination coefficients of the first order, Grau and Stover-Kincanon models for MBBR and SBR were obtained to be 0.710, 0.478 and 0.953 and 0.454, 0.692 and 0.972, respectively.

Microbial culture and gram staining results

According to the images taken from lamella, the existence of coccus and bacillus bacteria on the microbial culture were confirmed. Both gram positive and negative coccus and bacillus and gram positive binary bacillus were observed (Fig. 19).

DISCUSSION

According to Fig. 4, both systems had complete removal efficiency up to formaldehyde and glucose COD concentration of 100 mg/L. 100% removal efficiency was obtained in SBR that was higher than MBBR (93%) for formaldehyde COD of 200 mg/L. MBBR ad SBR had the efficiencies

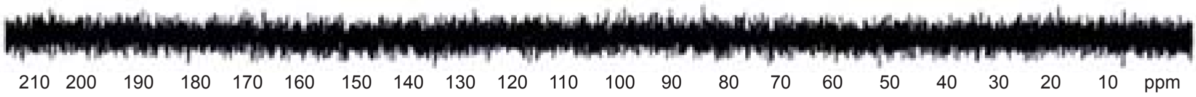

Fig. 12: CNMR spectroscopy result for the effluent sample

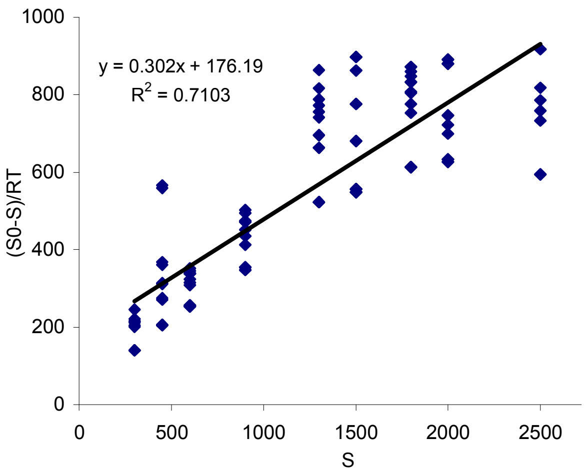

Fig. 13: First order model for MBBR

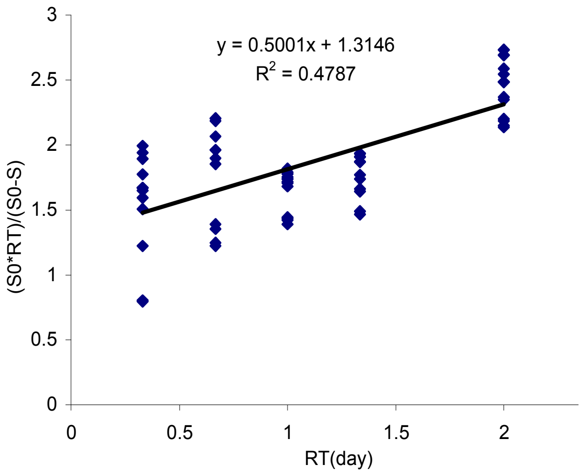

Fig. 14: Grau model for MBBR

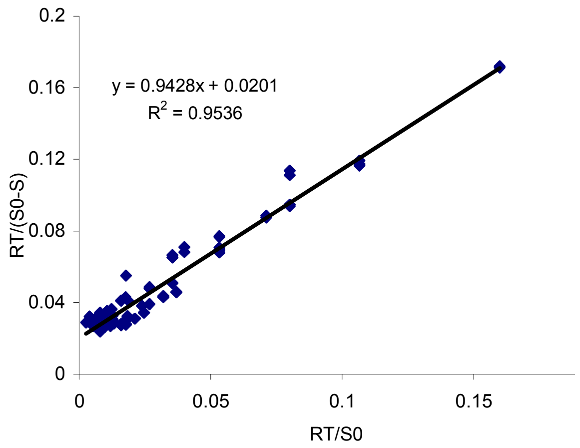

Fig. 15: Stover-Kincanon model for MBBR

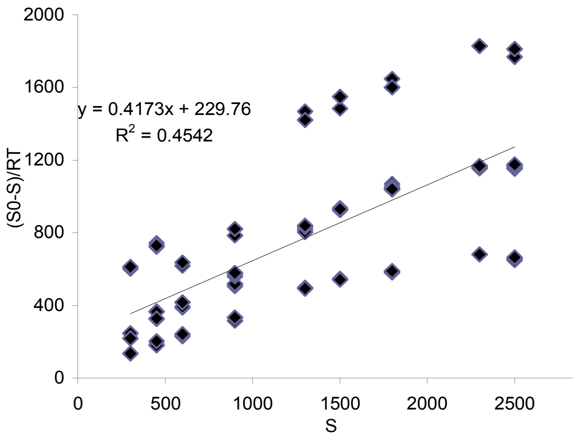

Fig. 16: First order model for SBR

less than 80% for the influent COD of more than 1500 and 1800 mg/L (in retention time of 48 hours), respectively.

Kashefiolasl and Nikkhah (2003) have reported that using activated sludge process in treatment of formaldehyde wastewater with initial concentration of 450 mg/L, 45% of COD has been removed. Whereas in this study using MBBR and SBR with the same initial oncentration resulted in 91.6% and 96.75% removal, respectively.

In this research (Figs. 5 and 6), different behaviors for variations of influent formaldehyde COD were observed. The removal efficiency of COD up to 450 mg/L (0.225 g COD/L.d) was reduced 2-4% for each 30% increase in COD, whereas for influent COD of higher than 1300 mg/L (0.65 g COD/L.d), the efficiency was reduced 7-13% by the same amount of increase in the influent COD. In COD between 450 to 1300 mg/L, the MBBR and SBR efficiencies were approximately

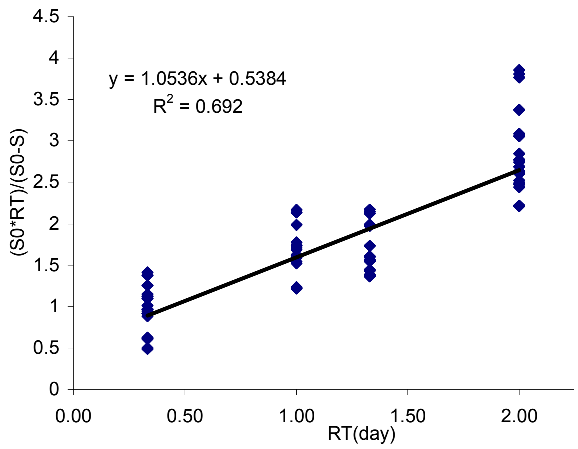

Fig. 17: Grau model for SBR

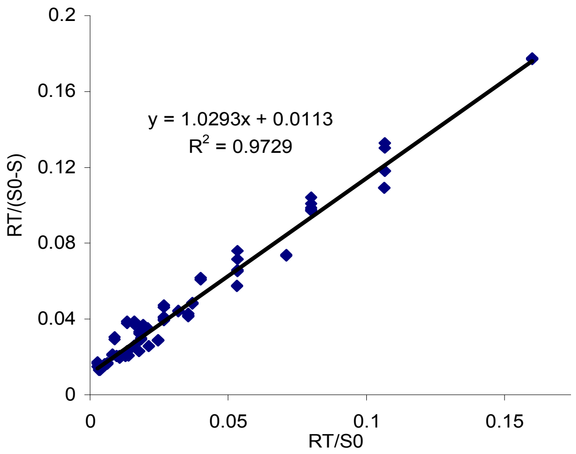

Fig. 18: Stover-Kincanon model for SBR

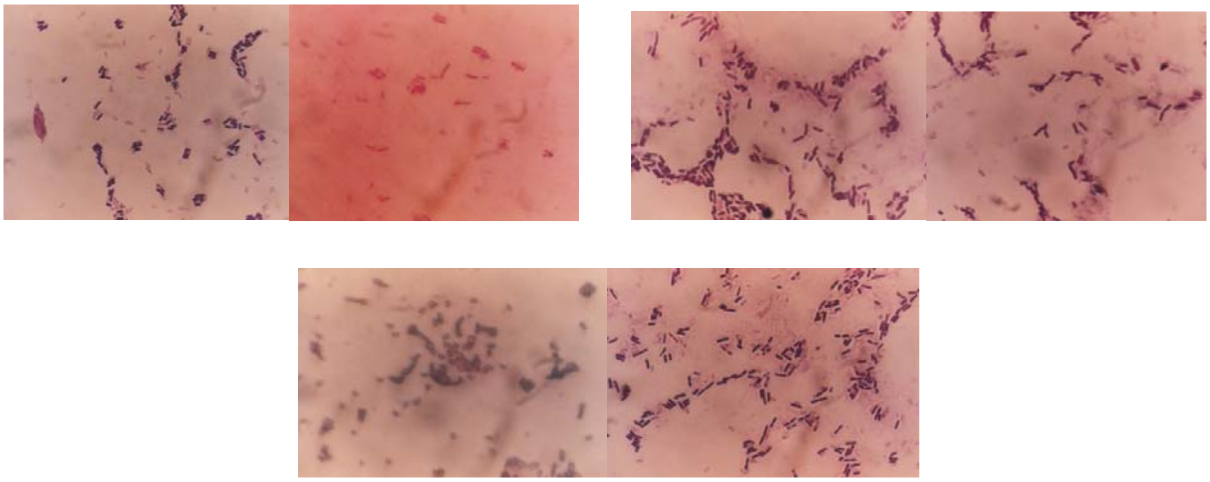

Fig. 19: The results attained from the photography of lamella

constant.

In COD of less than 450 mg/L, efficiencies differences between SBR and MBBR were less than 6%. In the influent substrates between 450 to 1300 mg/L, this amount was between 6 to 10 percent and in the COD of more than 1300 mg/L, it increased to 15% (Fig. 7).

It can also be seen that SBR had higher efficiency than MBBR. But according to t-test analysis, in the significance level of 0.05, the difference between COD removal of two systems was not significant.

MBBR and SBR systems can be used as pretreatment units for treating formaldehyde COD concentrations up to 2500 mg/L. They have shown an efficiency of 47 and 62 percent, respectively.

As shown in Fig. 8, the effect of media volume decrease is in balance with increasing in the retention time. That is, during 48-hour retention time, the MBBR with 30, 50, and 70 percent media filling ratios reached to the removal efficiencies of 35, 41% and 47%, respectively.

As shown in Figs. 9 and 10, at first shock loading on pilots, the efficiencies of aerobic systems decreased extensively whereas efficiencies of both MBBR and SBR gradually attained a stable situation after 14 and 23 days, respectively. Therefore, MBBR system seems to be more vulnerable to shock as compared with SBR system.

According to Fig. 11, *N*-{[(amino carbonyl) amino] methyl} urea was achieved in the influent samples from the reaction of formaldehyde and urea and 1, 3, 5-trioxane was produced from polymerization of formaldehyde residuals. It can be seen from Fig. 12 that all molecules containing carbon were degraded and there was no carbon in the samples.

In a previous study using CNMR spectroscopy, the possibility of formaldehyde and methanol biological degradation has been confirmed (Amato *et al.*, 2007(. In this research, CNMR spectroscopy has also confirmed the possibility of formaldehyde removal by aerobic biological treatment.

According to Fig.s 13-18, Stover-Kincanon model has fitted to the formaldehyde biodegradation data very well and this is confirmed by previous researches (Priya *et. al*, 2008).

According to Fig. 19, formaldehyde treatment by existing bacteria in the treatment plant after adaptation period was possible. It was also confirmed the findings of previous researches in this context (WHO, 1989).

In many studies, the ability of special microbial species in formaldehyde treatment has been investigated (Ladhari *et al.*,2005; Shinagawa *et al.*, 2008). Based on the results of this study which are obtained from microbial culture and photographing stained lamella, after adaptation period, bacillus and coccus species of domestic wastewater treatment plant can remove formaldehyde effectively.

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