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Feasibility Study on EO/EG Wastewater Treatment Using Pilot Scale SBR

Shakerkhatibi, M.[1*], Monajemi, P.[2], Jafarzadeh, M.T.[3], Mokhtari, S.A.[4] and Farshchian, M.R.[1]

[1]Department of Environmental Health Engineering, Tabriz University of Medical Sciences, Tabriz, Iran

[2]Department of Civil Engineering, University of Shiraz, Shiraz, Iran

[3]Manager of Environment, National Iranian Petrochemical Company, Tehran, Iran

[4]Department of Environmental Health Engineering, Ardabil University of Medical Sciences, Ardabil, Iran

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ABSTRACT: The sequencing batch reactors were developed to treat the actual wastewater from ethylene glycol/ethylene oxide producing industry. Four identical reactors with total and effective liquid volumes of 9 and 7 L were operated respectively in parallel. Laboratory experiments were conducted with different organic loadings of 500, 1000, 1500 and 3000 g-COD/m^3.day and the performance of the reactors was studied under different sludge ages of 10, 20 and 30 days, the kinetic constants at optimal operational conditions were also determined. According to the results, the efficient removal of COD was 79.5 and 83.5% (SRT = 20 days) and 86% (SRT = 30 days) for SBR 1 and 2 respectively (OLR = 0.5 to 1 kg-COD/m^3.day) with required reaction time of about 17 h. In order to reach the COD removal efficiencies over 86% at higher OLR values, the researchers required longer reaction periods about 34.5 h for SBR 3 and 4. However, the SRT values have no significant effects on the performance of SBR 1 and 2 at the aeration times greater than 22.5 h while in the case of reactors 3 and 4, with an increase on SRT at all aeration times, the COD removal efficiency increased. It is concluded that the system used in the present study could show an acceptable stability and performance in the treatment of the wastewater containing high concentrations of organic matters especially from EO/EG industries.

Key words: Petrochemical wastewater, SBR, Ethylene Glycol/Ethylene Oxide, Kinetic coefficients

INTRODUCTION

It has become increasingly important to prevent the pollution of the limited water resources by providing adequate treatment of the effluents from industrial sources (Nabi Bidhendi *et al.*, 2010; Mirbagheri *et al*., 2010; Usharani *et al.*, 2012; Uemura *et al.*, 2012; Mobarak-Qamsari *et al.*, 2012; Okuku *et al.*, 2011; Khurram, 2011; Houda *et al.*, 2011; Haruna *et al.*, 2011; Naim *et al.*, 2011). One of the major environmental problems in Iran is the disposal of wastes generated by industries. Petrochemical industry can be considered as one of the most important industries which were exposed to a great development due to specific-economic conditions of the country and increasing demand for petrochemical products during the recent years. Petrochemical activities result in many problems of which high environmental pollution is the most important one that can lead to several direct effects on social and environmental health and almost appears in three dimensions of water, soil and vibrations (Malmasi *et al.*, 2010; Mirbagheri *et al*., 2010; Rahman and Al-Malack, 2012; Hatamoto *et al.*, 2012; Arshad *et al.*, 2011).

Wastewaters generated from petrochemical processes contain a large number of pollutants at high concentrations and have adverse environmental impacts (Malmasi *et al.*, 2010; Papadimitriou *et al*., 2009). A certain type of wastewaters, such as those produced from Ethylene Oxide/Ethylene Glycol (EO/EG) plants; contain high chemical oxygen demand (COD) in accordance with mono ethylene glycol and its derivatives as well as aldehydes. The concentration of pollutants may reach up to 20000 mg/L as COD and 90 mg/L as aldehydes, respectively.

In Iran, wastewater in petrochemical industry is currently treated with conventional activated sludge process (CASP). It has been successfully used for

*Corresponding author E-mail: shakerkhatibim@tbzmed.ac.ir

treating petrochemical wastewaters so the treated effluent usually meets the industrial wastewater discharge standards (Fallah *et al*., 2010). However, there are some common problems associated with conventional methods for treating the waste streams with special characteristics e.g. for the wastewater generated from EO/EG, acrylonitrile-butadiene-styrene (ABS) (Shakerkhatibi *et al.*, 2010) and some other effluents from petrochemical industries. For instance, the introduction of EO/EG effluents to the central CASP located at Arak Petrochemical Complex can result in the loss of the system performance due to its high COD concentrations. Therefore, finding an alternative method for treating EO/EG wastewater has become a critical issue to satisfy the effluent discharge standards. Increasingly, the sequencing batch reactor (SBR) has been demonstrated to be a viable alternative to the conventional continuous-flow activated sludge process (Chan and Lim, 2007; Mahmoudkhani *et al.*, 2012; Selvamurugan *et al.*, 2012; Adl *et al.*, 2012; Uemura *et al.*, 2011). It is known as a flexible and effective system for the biological treatment of wastewater, even though there are high concentrations of toxic compounds produced by various industrial processes. An SBR is a periodically operated, fill-and-draw reactor. It has five discrete periods in each operation cycle including fill, react, settle, draw, and idle. Reactions start during fill and complete during react. After react, the mixed liquor suspended solids (MLSS) are allowed to separate by sedimentation during settle in a defined time period; the treated effluent is withdrawn during draw. The time period between the end of the draw and the beginning of the new fill is termed idle (Tsang *et al.*, 2007; Malakahmad *et al.*, 2011). A number of papers which provide good description and evaluation of the SBR systems in COD, nitrogen, and phosphorous removal (Kim *et al.*, 2008), treatment of petrochemical wastewater containing Hg and Cd (Malakahmad *et al.*, 2011), textile wastewater treatment and reclamation (Khouni *et al.*, 2012; Zuriaga Agusti *et al.*, 2010), treatment of high nitrogen loaded wastewater (Ganigue *et al.*, 2012) and treatment of different industrial wastewaters (Val del Rio *et al.*, 2012; Kulkarni, 2012; Volmajer Valh *et al.*, 2012; Mwinyihija, 2012; Onodera *et al.*, 2012; Arshad and Hashim, 2012; Takahashi *et al.*, 2011; Hassani *et al.*, 2011; Han *et al.*, 2011; Safari *et al.*, 2011; Amani *et al.*, 2011; Akbarpour Toloti and Mehrdadi, 2011) have been published. However, there are limited investigations in the literature on the treatment of the EO/EG containing petrochemical wastewater using SBR technology. The wastewater generated from such industries constitute various organic substances used in the process, inorganic salts, organic solvents, etc. which result in high COD, low BOD, high salt content (TDS), toxic and inhibitory substances in wastewater which inhibit the biological process (Mohan *et al.*, 2005).

The purpose of the present study was to investigate the application of SBR technology for treating EO/EG plant effluents at the Arak petrochemical complex located in the center of Iran. In addition, the influence of the varying of the organic loading rates (OLRs), the hydraulic retention times (HRTs) and sludge age (SRT) on the reactor performance were studied. This work also aims to determine the kinetic coefficients based on Monod kinetics.

MATERIALS & METHODS

Four identical plexi glass reactors with total and effective liquid volumes of 9 and 7 L were operated respectively in parallel. The reactors had an internal diameter of 0.2 m and 0.3 m length with the outlet of the reactor used for wastewater withdrawal at 0.09 m length from the bottom of the reactor. Air supply was by means of diffused aerators placed at the bottom of the reactors connected to a sparger arrangement. The amount of oxygen supplied in the aeration phase was regulated in order to maintain oxygen concentration over 2 mg/L in all the reactors during the experiments. Feeding, sludge wasting and effluent discharge were performed by peristaltic pumps (RP-1000 EYELA). The rough openings located in the cover of the reactors. Aerators, feed and decant pumps were controlled by timers. Mixing was ensured by a magnetic stirrer. At the beginning of each cycle, immediately after withdrawal, a pre-defined feed volume was pumped into the system and the reactor volume was re-circulated with aeration during the reaction phase. At the end of the cycle, suspended biomass settled and effluent was withdrawn from the reactor. During each cycle, 3.5 L of the feed solution were introduced continuously during the FILL period and the same volume of treated effluent was removed during the DRAW period. The pH of the influent was adjusted to 7.1±0.2 before wastewater feeding. The lay-out of the pilot-scale reactor and control system are shown in Fig. 1.

Two series of experiments were carried out in the pilot-scale SBRs. In the first one, for the start-up of the reactors, the researchers collected the activated sludge used as inoculums from the return sludge of the central wastewater treatment plant in Arak petrochemical complex. The collected sludge was aerated for 1–2 days at room temperature before inoculating the reactors. Then, the sludge was introduced to SBRs in order to obtain a concentration of total suspended solids of around 5 g/L. During the start-up of the reactors, Hydraulic Residence Times (HRTs) of 24 h and Sludge Residence Times (SRTs) of 11.7 days were employed. Biomass concentration in the reactors was maintained

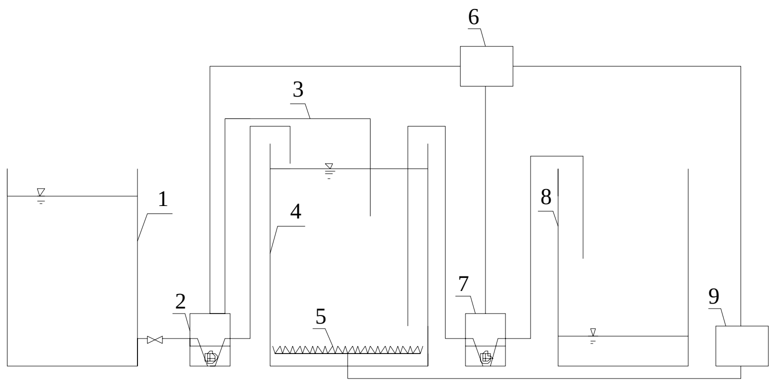

Fig. 1. Schematic representation of the sequencing batch reactor.
1. Feed tank; 2. Peristaltic pump (Feed); 3. Level sensor; 4. SBR reactor; 5. Air diffuser; 6. Timer; 7. Peristaltic pump (Effluent); 8. Effluent tank; 9. Air compressor

between 3500 and 5000 mg-VSS/L. The system had been operated at the room temperature (20°C) during the first month of experiments to reach a steady state before starting further operations. At the end of the start-up period, the OLRs in SBR 1 to 4 reached 500, 1000, 1500 and 3000 g-COD/m^3.day respectively.

In the second series of experiments, 3 runs were carried out with different SRT so that in the 1st, 2nd and 3rd runs, the sludge ages were 10, 20 and 30 days respectively. The SRT in this runs was maintained by carrying off the excess sludge in the amounts of 350 cm^3/cycle in the 1^{st} run, 175 cm^3/cycle in the 2^{nd} run and 115 cm^3/cycle in the 3^{rd} run. In all the runs, the HRT were 0.5 day (SBR 1), 0.75 day (SBR 2), 1 day (SBR 3) and 1.5 days (SBR 4). The effluent from the EO/EG industry located at Arak petrochemical complex used as the influent of SBRs, was collected and immediately refrigerated at 4°C. Nutrients such as urea and di-potassium hydrogen phosphate were supplied to maintain a COD:N:P ratio of 100:5:1. In all the runs, the actual wastewater from EO/EG industry was introduced to the SBR 1, 2, 3 and 4 with the COD concentrations of 1000, 2000, 3000 and 6000 mg/L respectively. The detailed characteristics of the wastewater from EO/EG industry in Arak petrochemical complex are given in Table 1. The phase duration and operating conditions of the SBRs are shown in Table 2. The reactors were operated for at least 2 weeks at each combination of operating conditions investigated in order to ensure stable performance before the corresponding data collection. The performance of the reactors was evaluated at the influent of COD concentrations and different operating conditions of each cycle. After the attainment of stable operating conditions in terms of the COD treatment efficiency and the mixed liquor suspended solids (MLSS) concentration for each influent COD concentration, the major parameters were determined.

During the study, the concentration of influent and effluent COD (Standard code: 5220), pH (Martini instruments, pH 56), the dissolved oxygen (Eutech instruments, Cyberscan DO 300), TSS (Standard code: 2540-D), VSS (Standard code: 2540-E), NH_4^+-N (Standard code: 4500-NH_3-D), NO_3^--N (Standard code: 4500-NO_3^--B), PO_4^{3-}-P (Standard code: 4500) and alkalinity (Standard code: 2320) were measured. The analytical procedures used for parameter determination were those outlined in the Standard Methods (APHA, 2005). All the soluble parameters were determined in samples filtered through 0.45 μm membrane filter.

RESULTS & DISCUSSION

The start-up period of the reactors was carried out using glucose as biogenic substrate with the influent COD concentrations of 1000, 2000, 3000 and 6000 mg/L for SBR 1 to 4 respectively. During the acclimation period, glucose was replaced with the actual EO/EG effluent gradually, to reach the desired concentrations. Fig. 2 presents the COD removal efficiency of SBRs at different concentrations of feed wastewater from 17 to 100 percent of which wastewater with 100% concentration refers the mean COD values of 6000 mg/L. As can be seen from Fig. 2, all reactors presented an efficient performance for pollutant removal so the COD removal efficiencies were almost the same at all reactors. The most striking result to emerge from the data presented in Fig. 2 is that a significant portion of the influent COD is biologically degradable.

After starting-up of the reactors, the SBRs were operated under variable aeration time, HRTs and sludge ages. All the runs were continued to achieve steady state conditions and the reactors performance was

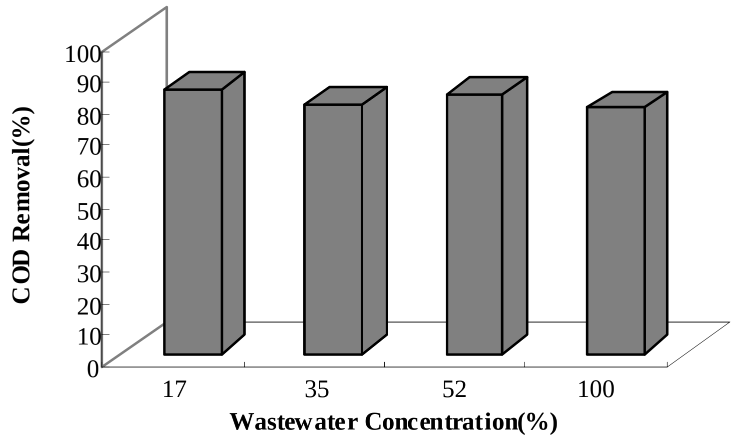

Fig. 2. COD removal during start-up period at HRT of 24 h.

assessed by monitoring COD removal during the sequences operation and also throughout the reactors operation. The reactors were assumed to be operating at steady state when the measured effluent COD concentrations did not show variations higher than 10% at least for the last five measurements.

The effect of aeration time on the performance of reactors, such as COD removal, was studied at four different time of aeration including 10.5, 16.5, 22.5 and 34.5 h with the sludge ages of 10, 20 and 30 days. The variation of COD removal as a function of aeration time under sludge ages of 20 and 30 days are depicted in Fig. 3 and 4 respectively.

As shown in Figs. 3 and 4, SBR 1 and 2 required about 17 h of the react period to remove the significant portion of the COD. Accordingly, 79.5 and 83.5 percent COD removal at SRT of 20 days and 86 percent COD removal at SRT of 30 days were observed respectively for SBR 1 and 2 at the OLRs of 0.5 and 1 kg-COD/m^3.day. However, the COD removal rates were slow at the aeration time greater than 17 h for the reactors 1 and 2. To reach the COD removal of 86% at the SRT of 20 days and 92% at the SRT of 30 days, the aeration time of 34.5 h was required at the OLR of 1 kg-COD/m^3.day for SBR 3. Another important finding was that the aeration times required for SBR 4 were 34.5 and 22.5 h to reach the COD removal of 89.7 and 91.1% at the sludge ages of 20 and 30 days respectively. According to the results, the SRT has no significant effects on the performance of SBR 1 and 2 at the aeration time greater than 22.5 h. Although, these results differ from some published studies i.e. (Sirianuntapiboon and Boonchupleing, 2009), they are consistent with Karapinar Kapdan and Ozturk (2005) who found that the increasing in SRT from 15 to 30 days did not affect the color and COD removal efficiencies of SBR treating an industrial wastewater. This may be due to the limited capacity of microorganisms and/or it may also be the result of the limitation of oxygen transfer within the SBR 1 and 2. However, in the case of reactors 3 and 4, with an increase on SRT at the all aeration time, the COD removal efficiencies were increased. It is encouraging to compare this finding with that found by Sirianuntapiboon and Boonchupleing (2009) who found that the removal efficiencies of SBR system treating industrial wastewater were increased with the increase of MLSS concentrations.

Initially, it seemed that the appropriate sludge age for the SBR was about 10 days, while the results showed that the SRT value of 10 days was insufficient especially at the longer HRTs such as 24 and 36 h which can reduce the sludge content in the system gradually. The effect of the sludge age on the SBRs performance at the HRT values of 24 and 36 h is shown in Figs. 5 and 6 respectively. As shown in Figs. 5 and 6, the COD removal rate is significantly affected by the sludge age, so the difference between COD removal efficiency at the SRT of 30 versus 10 days is remarkable. However, it seems that the appropriate SRT values for influent COD concentrations of 1000 to 2000 mg/L and 3000 to 6000 mg/L were about 20 and 30 days respectively. It is apparent from these Figs. that the decrease in the amount of the sludge in the system caused a significant decrease in the elimination of the COD. This finding is in agreement with Sirianuntapiboon and Boonchupleing's (2009) findings which showed the direct relationship between bio-sludge concentrations and the COD removal efficiencies in SBR.

The effect of SRT on the MLSS concentrations under HRT values of 24 and 36 h is shown in Figs. 7 and 8 respectively. As it can be seen from the Figs. 7 and 8, there was no significant difference in MLSS concentrations at the HRT values of 24 and 36 h, however the MLSS concentrations at all studied HRTs were affected by SRT values (Sirianuntapiboon and

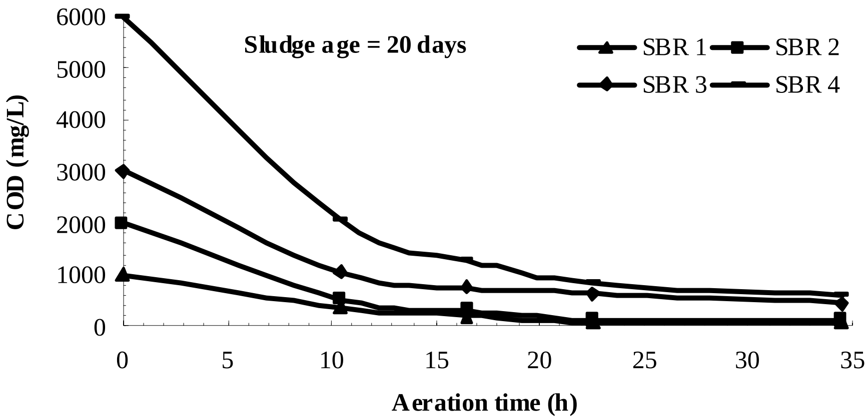

Fig. 3. Variation of COD as a function of aeration time at the SRT of 20 days

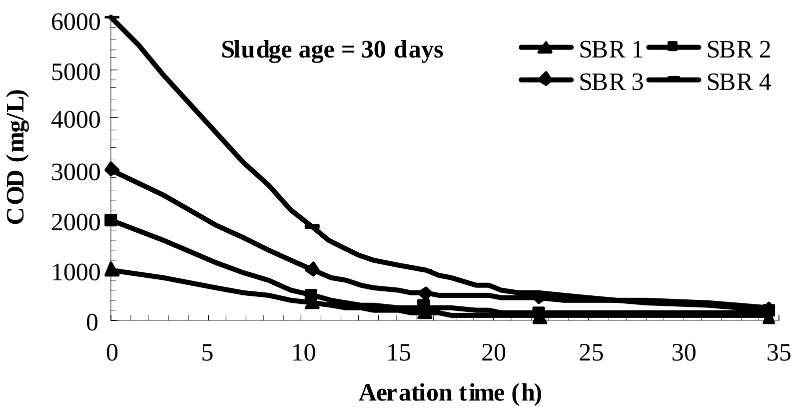

Fig. 4. Variation of COD as a function of aeration time at the SRT of 30 days

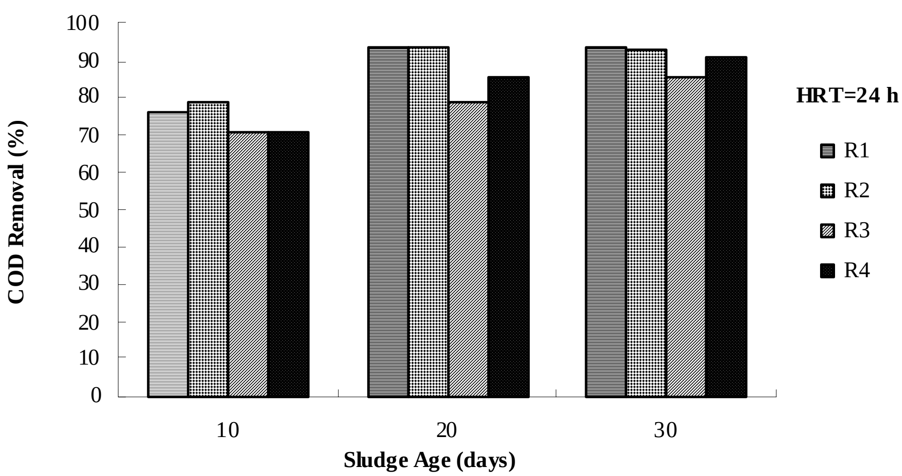

Fig. 5. COD removal efficiencies under various SRT at the HRT of 24 h.

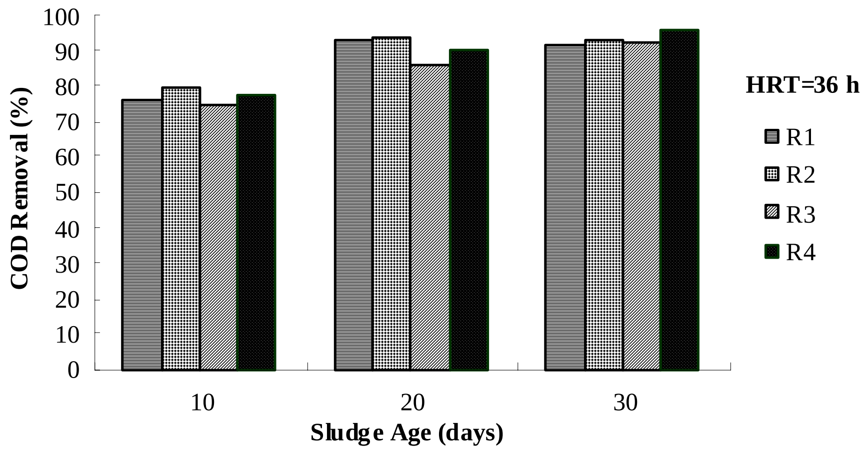

Fig. 6. COD removal efficiencies under various SRT at the HRT of 36 h

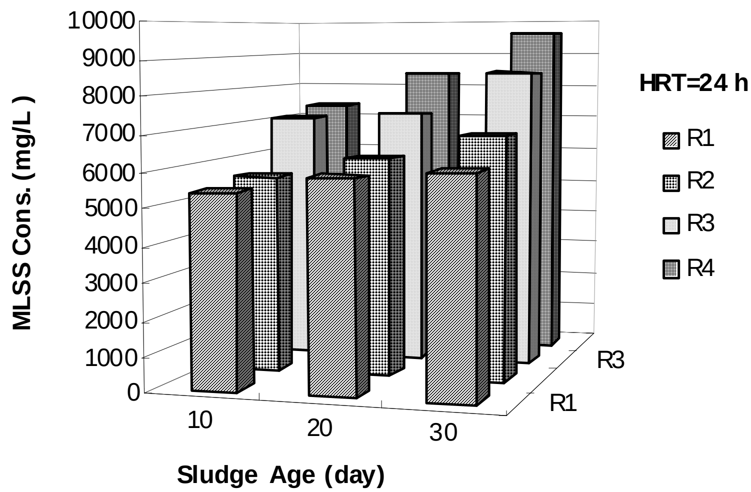

Fig. 7. MLSS concentrations under various SRTs at the HRT of 24 h

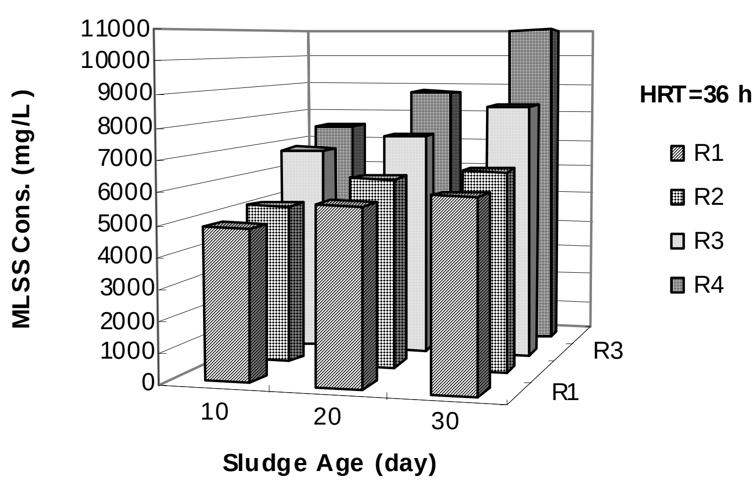

Fig. 8. MLSS concentrations under various SRTs at the HRT of 36 h

Boonchupleing, 2009). In addition, comparing the MLSS concentrations between the two systems of SBR and the conventional activated sludge revealed that the values observed in this study were greater than those reported in the literature for the conventional activated sludge systems.

Fig. 9 provides the results of sludge settling experiments at different HRTs. No significant differences were found between the curves obtained at the HRTs of 36 and 48 h. However, the elapsed time for settling of the sludge increased when the HRT values decreased. In other words, the sludge generated at higher HRT conditions had better settling characteristics due to formation of heavier flocs. This finding supports previous research into this area which links HRT and sludge settling characteristics (Sirianuntapiboon *et al.*, 2007). Fig. 10 compares the optimum time for sludge settling at different HRTs. The data in Fig. 10 depicts the length of time to obtain stable settling is longer when the SBR is operated under shorter HRTs. The results showed that the optimum settling time were 70 and 50 min at the HRT values of 24 and 36 h respectively.

One of the most important parameters to assess the stability of the sludge in any aerobic suspended growth system is sludge volume index (SVI). SVI was monitored during the experiments at the end of each cycle before starting decant period. The results obtained from the determination of SVI at the steady state conditions under different values of HRT are presented in Table 3. The SVI was relatively high initially at the OLR of 0.5 kg-COD/m^3.day with corresponded HRT of 18 h. An increase of OLR resulted in a decrease of SVI. However, the SVI was good (below 100) during the stable operation of SBRs. As mentioned in the literature by Mohan *et al*., (2005) the sludge normally settles well in SBR at high concentration of DO, in comparison with other aerobic suspended growth systems. During all experimental runs in present study, the concentration of DO was maintained at the range of 2 to 4 mg/L.

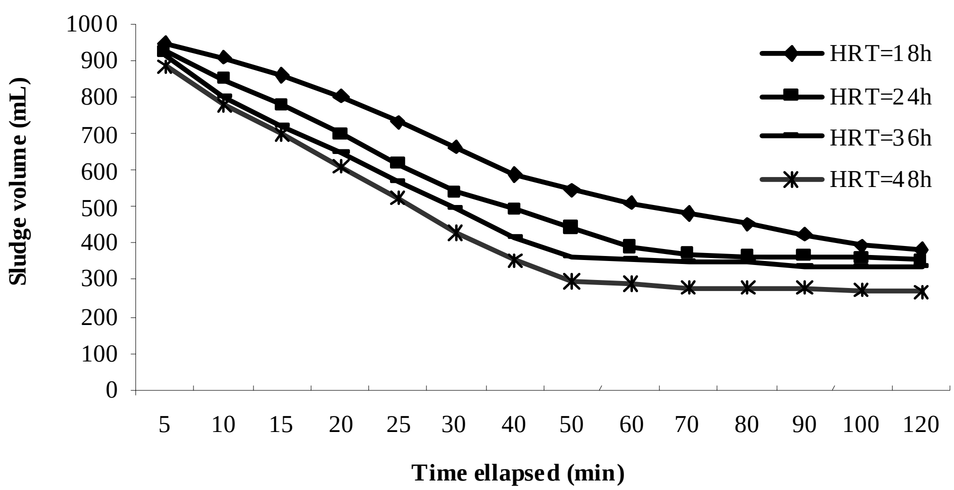

Fig. 9. Settling curves at various HRTs

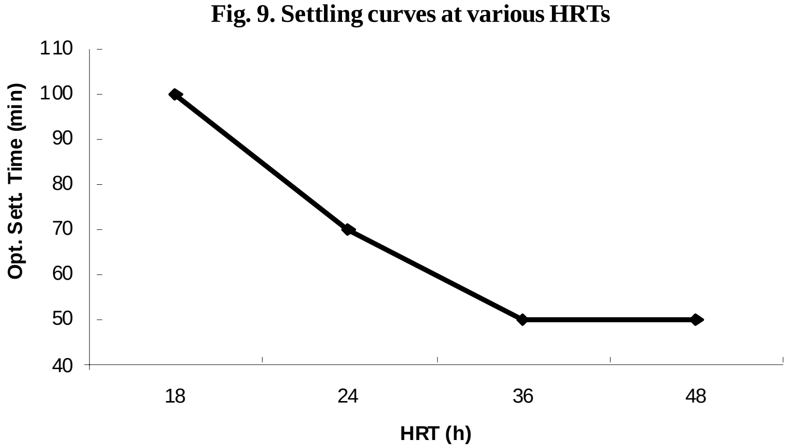

Fig. 10. Optimum settling times as a function of HRT in SBRs (Sludge age: 30 days)

The performance of SBR 1 to 4 as COD removal efficiency at various operational conditions in terms of aeration period, HRT and SRT was compared in Table 4. According to the data in this table the most acceptable removal efficiencies of 93.6% were obtained for SBR 1 (OLR = 0.5 kg-COD/m^3.day) and SBR 2 (OLR = 1 kg-COD/m^3.day) at the HRT and SRT values of 24 h and 20 days respectively. However, in order to reach the highest removal efficiencies for SBR 3 (OLR = 1 kg-COD/m^3.day) and 4 (OLR = 2 kg-COD/m^3.day), longer HRT and SRT of 36 h and 30 days were needed, respectively.

The results of experimental analysis in terms of MLSS and effluent COD concentrations at various loading rates in all reactors as well as the calculated parameters for determining kinetic coefficients are presented in Table 5. Table 5 indicates that the MLSS concentrations for SBR 1 to 4 increased as the aeration time increased except for the ones whose time is longer than 24 h. However, the MLSS values for reactor 4 were the highest and for the reactor 1 were lower than the others. This finding is in agreement with Chan and Lim's (2007) findings that showed an increase in MLSS concentrations due to an increase in the influent substrate concentrations in two SBR configurations. The linear regression for determination of Y and k_d during the reaction periods of 22 and 34 h are shown in Figs. 11 and 12 respectively. These illustrations show that the values of Y were 0.433 and 0.509 mg-MLSS/mg-COD and the values of k_d were 0.0647 and 0.0652 1/day respectively. It was observed that the Y value for reaction period of 34 h was relatively higher than the reaction period of 22 h. However, the values of k_d in both reaction time were almost the same.

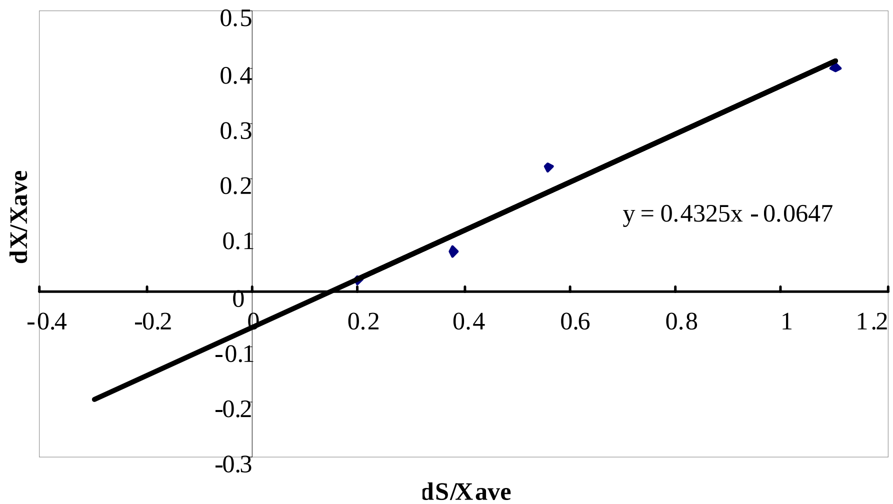

Fig. 11. Linear regression for determination of Y and k_d (reaction time = 22 h)

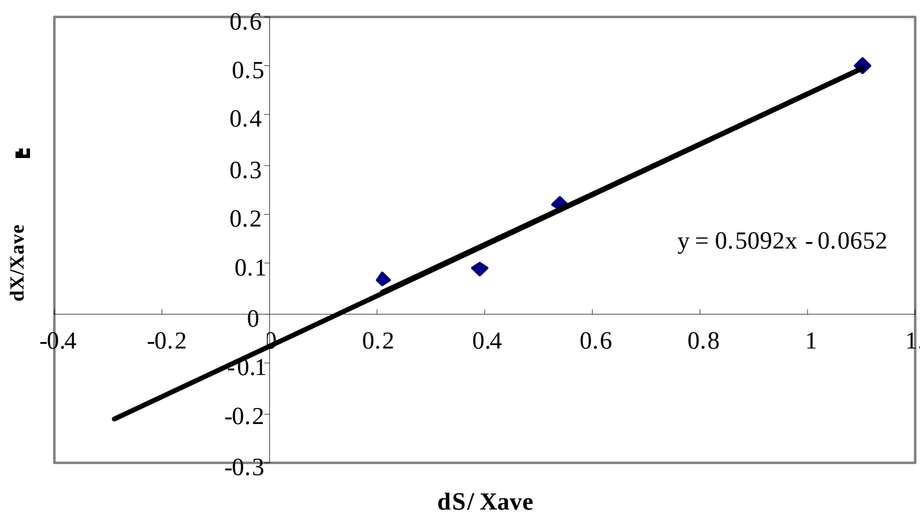

Fig. 12. Linear regression for determination of Y and k_d (reaction time = 34 h)

CONCLUSION

This study aimed to investigate the application of SBR technology for treating petrochemical effluents containing EO/EG. During the five-month experiments, the stable performance of SBRs was achieved. The following was drawn from the obtained results:

-A small fraction of constituents found in EO/EG containing wastewater was not easily biodegradable at the start up period.

-With increase in hydraulic retention time (up to 24 h for SBR 1 and 2 and up to 36 h for SBR 3 and 4), the COD removal rate increased ;however, the performance of the reactors was not significantly improved at higher HRTs.

-The effluent COD from SBRs is dependent on sludge age and the optimum SRT at the influent COD of 1000-2000 mg/L and 3000-6000 mg/L were 20 and 30 days respectively.

-At the shorter hydraulic retention time, longer time is needed for the stable settling so the optimum sludge settling time was observed 70 and 50 min at corresponding HRTs of 24 and 36 h, respectively.

-During nearly all experimental runs, the SVI values were at the range of 50-100 indicating good settling characteristics.

These results support the applicability of sequencing batch reactors to full-scale treatment of EO/EG containing petrochemical wastewater.

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