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Optimization of Sequencing Batch Reactor Operating Conditions for Treatment of High-strength Pharmaceutical Wastewater

[1]Emad S. Elmolla, [2]Natasha Ramdass and [2]Malay Chaudhuri
[1]Department of Civil Engineering, Faculty of Engineering, Al-Azhar University, Cairo, Egypt
[2]Department of Civil Engineering, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, 31750 Tronoh, Perak, Malaysia

Corresponding Author: Emad S. Elmolla, Department of Civil Engineering, Faculty of Engineering, Al-Azhar University, Cairo, Egypt

ABSTRACT

Optimization of the Sequencing Batch Reactor (SBR) operating conditions for high-strength non-penicillin pharmaceutical wastewater treatment is a challenging task. Two SBR were operated under different operating conditions. Three different Hydraulic Retention Times (HRT) (12, 24 and 48 h) were tested and operated under high (6000 mg L^{-1}) and low (4000 mg L^{-1}) Mixed Liquor Suspended Solids (MLSS) concentration. Statistical analysis (two-way ANOVA) using SPSS software was applied on the results to evaluate the effect of HRT and MLSS concentration on the SBR performance. The hydraulic retention time of 24 h was found suitable for the SBR and increasing HRT from 12 to 24 h significantly affected the BOD_5 removal; however, increasing HRT from 24 to 48 h or increasing MLSS concentration from 4000 to 6000 mg L^{-1} did not significantly improve the SBR performance in terms of BOD_5 removal. Under the optimum operating conditions (HRT 24 h and MLSS 4000 mg L^{-1}), the SBR achieved an efficiency of 94±1.3% in terms of BOD_5 removal and 83.9±1.7% in terms of COD removal, with complete nitrification.

Key words: Non-penicillin pharmaceutical wastewater, optimization, sequencing batch reactor

INTRODUCTION

Pharmaceutical compounds have been detected in surface water (Kolpin *et al.*, 2002; Anderson *et al.*, 2004; Rabiet *et al.*, 2006), ground water (Rabiet *et al.*, 2006), sewage effluents (Carballa *et al.*, 2004; Nikolaou *et al.*, 2007) and even in drinking water (Stackelberg *et al.*, 2004). Pharmaceutical compounds can reach the aquatic environment through various sources such as pharmaceutical industry, hospital effluent and excretion from humans and livestock (Ikehata *et al.*, 2006; Nikolaou *et al.*, 2007; Yang *et al.*, 2008). Inorganic and organic raw materials may be used in the pharmaceutical industry and hence, the generated wastewater may be high in COD and Total Suspended Solids (TSS), with wide range of pH (Gotvajn and Zagorc-Koncan, 2003). Generally, most of the pharmaceutical wastewater is toxic to biological life and the biodegradability ratio (BOD/COD) is low (Oktem *et al.*, 2007).

Sequencing Batch Reactor (SBR) is a wastewater treatment system based on the principles of the activated sludge process. The wastewater is treated in batches with aeration and settlement both occurring in the same tank. The SBR process consists of several phases: filling,

aeration-reaction, settling, decant and idle phase. With respect to application, SBR has been successfully used in both municipal and industrial wastewater treatment (Mace and Mata-Alvarez, 2002).

SBR has many positive processing characteristics. For example, combining the reactor and the setting tank in the same vessel easily controls performance with respect to reaction time and sludge solids maintenance. Aeration of the mixed liquor occurs in the first two stages. Sludge is formed and ammonia is oxidized to nitrites and nitrates. In the settling stage, settlement of the sludge takes place and denitrification process may occur due to continuous consumption of nitrogen (Neczaj *et al*., 2008). The SBR process is comparatively easy to operate and the process saves more than 60% of the expenses when compared with the conventional activated sludge process (Chang *et al*., 2000).

SBR is known as an effective biological treatment system for treatment of domestic wastewater (Mace and Mata-Alvarez, 2002). In addition, it has been reported as an effective biological treatment system for many of industrial wastewater such as landfill leachate (Neczaj *et al*., 2008); pulp and paper industry wastewater (Tsang *et al*., 2007); dairy wastewater (Neczaj *et al*., 2008) and chemical complex wastewater (Mohan *et al*., 2005), photo-Fenton treated antibiotic wastewater (Elmolla and Chaudhuri, 2011).

The objective of this study was to determine the optimum operating conditions for effective treatment of a high-strength non-penicillin pharmaceutical wastewater by SBR as well as study the effect of HRT and MLSS concentration on the SBR performance.

MATERIALS AND METHODS

Pharmaceutical wastewater: The pharmaceutical wastewater was collected from a pharmaceutical company in Bangi, Kuala Lumpur, Malaysia. The pharmaceutical wastewater was transported to the laboratory and stored at 4°C. Before taking a sample and starting the experiment, the wastewater was mixed well and left for 2 h to reach the room temperature. The characteristics of the pharmaceutical wastewater are summarized in Table 1.

Analytical methods: Chemical Oxygen Demand (COD) and five-day Biochemical Oxygen Demand (BOD_5), Total Suspended Solids (TSS), ammonia-nitrogen (NH_3-N), nitrate nitrogen (NO_3^--N) and Total Phosphorous (TP) were monitored throughout the operation. A pH meter (HACH Sension 4) with a pH electrode (HACH platinum series pH electrode model 51910, HACH Company, USA) was used for pH measurement. The pH meter was calibrated with pH 4.0, 7.0 and 10.0 buffers. COD, BOD_5, TSS, Mixed Liquor Suspended Solids (MLSS) and Mixed Liquor Volatile Suspended Solids (MLVSS) were analyzed according to the standard methods (APHA, AWWA and

Table 1: Characteristics of the pharmaceutical wastewater sample

Parameter	Concentration (mg L^{-1})
BOD_5	765±25
COD	1352±40
TSS	71.3±12
NH_3-N	6.8±2
NO_3^--N	1.8±0.5
Total phosphorus	18.1±2.5
Sulphate	20
Sulphide	0.28
pH	4.36

WEF, 1992). NH_3^--N was measured by the Nessler method (Method 8038), NO_3^--N by the cadmium reduction method (high range) using HACH powder pillow and TP by PhosVer 3 method using HACH powder pillow (HACH, 2003). Daily analyses of COD and BOD_5 for both influent and effluent were carried out. Sulphate and sulphide were determined by ion chromatograph (Metrohm). The eluent phase consisted of 3.2 mM Na_2CO_3 and 1.0 mM $NaHCO_3$. The analytical column was METROSEP A SUPP 5-150 (4.0×150 mm, 5 μm). The flow rate was 0.7 mL and the temperature was 20°C.

Experimental setup: A lab-scale setup was used in the study and it consisted of two identical SBR with operating volume of 1.5 L. The operating volume was divided into 1.0 L decanting volume and 0.5 L sludge volume. The reactor was equipped with an air pump and air diffuser to keep dissolved oxygen above 3 mg L^{-1} and magnetic stirrer for mixing purpose. Feeding and decanting were performed using two peristaltic pumps. The cycle period was divided into five phases: filling (0.25 h), aeration-reaction (variable), settling (1.25 h), decant (0.25 h) and idle (0.25 h). Cycle phases were controlled by an electric control panel. Figure 1 shows the experimental setup.

Startup of SBR: The seed sludge was obtained from the aeration tank of the Sewage Treatment Plant (STP) of the Universiti Teknologi Petronas campus. The two SBR were inoculated with different sludge concentration to get different MLSS concentration and in order to acclimate the biomass, HRT was chosen to be 2 days and the pharmaceutical wastewater was mixed with domestic wastewater obtained from the STP. The feed wastewater was a mix of the pharmaceutical wastewater and domestic wastewater at ratio of 25:75, 50:50, 75:25 and 100:0 and the acclimation period was extended to 20 days. Concentration of biomass (MLSS) after the acclimation period were 6000 and 4000 mg L^{-1} in SBR1 and SBR2, respectively.

Data analysis: Statistical analyses (ANOVA) was conducted using SPSS software and Microsoft excel. Two-way ANOVA was applied to the results to determine the significant difference between the data that were obtained for each variable of the experiments.

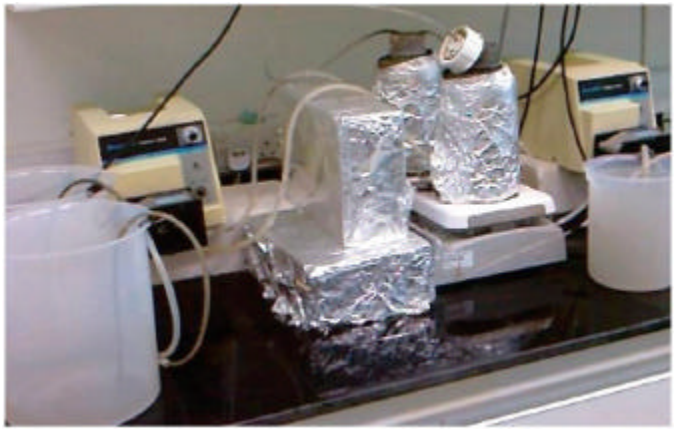

Fig. 1: Experimental setup

RESULTS AND DISCUSSION

Effect of MLSS concentration on the SBR performance: In order to study the effect of MLSS concentration on the SBR performance, SBR1 and SBR2 were operated in parallel. MLSS concentration in the SBR1 was 6000 mg L^{-1}; however, it was 4000 mg L^{-1} in the SBR2. The operating conditions such as pharmaceutical wastewater characteristics, temperature and SBR cycle period were kept unchanged for SBR1 and SBR2. The SBR cycle period was 24 h and it was divided into filling phase (0.25 h), aeration-reaction phase (22 h), settling phase (1.25 h), decanting phase (0.25 h) and idle phase (0.25 h). Figure 2 shows the effects of MLSS concentration on the SBR performance in terms of COD and BOD_5 removal. COD removal was 83.6±1.3 and 83.9±1.7%; however, BOD_5 removal was 93.7±1.4% and 94.0±1.3% in the SBR1 and SBR2, respectively. The results show that increasing MLSS concentration did not improve the SBR performance. The same finding has been reported in the literature (Tsang *et al.*, 2007). However, it disagrees with some other reported study on the effect of MLSS concentration on the conventional activated sludge process and up-flow aerated biofilter (Antonio *et al.*, 2003). The negative effects of the high MLSS concentration in the SBR could be ascribed to long sludge settling time and high concentration of suspended solids in the effluent (Tsang *et al.*, 2007). Based on the results, MLSS concentration of 4000 mg L^{-1} was considered suitable for treatment of the pharmaceutical wastewater by the SBR process.

Effect of HRT on the SBR performance: In order to study the effect of HRT on the SBR performance, three SBR cycle periods (12, 24 and 48 h) were studied. SBR1 was operated at MLSS concentration of 6000 mg L^{-1} and SBR2 at MLSS concentration of 4000 mg L^{-1}. The cycle period was divided to filling phase (0.25 h), settling phase (1.25 h), decanting phase (0.25 h) and idle phase (0.25 h) and the rest of cycle period was for aeration-reaction. Other operating conditions such as pharmaceutical wastewater characteristics and temperature were kept unchanged for SBR1 and SBR2. Figure 3 and 4 show the effects of cycle period (HRT) on the SBR1 performance in terms of COD removal (Fig. 3) and BOD_5 removal (Fig. 4).

It is important to know if increasing cycle period and MLSS concentration significantly affected the SBR performance. In order to do that, a two-way analysis of variance (ANOVA) was conducted using the SPSS statistical software. Table 2 shows the significance of the difference between the two means for COD removal in SBR1 and SBR2 under low (4000 mg L^{-1}) and high (6000 mg L^{-1}) MLSS concentration at HRT 12, 24 and 48 h, using the Tukey HSD method. When the value of significance is less than 0.05, it indicates that COD removal in SBR is significantly different

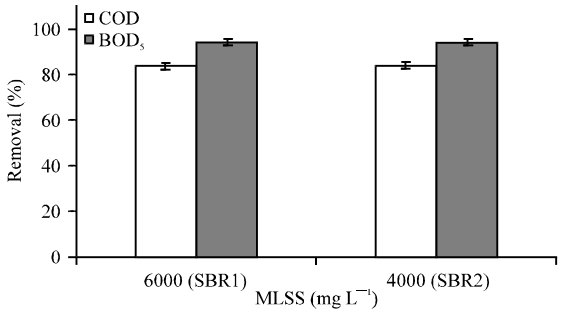

Fig. 2: Effect of MLSS concentrations on SBR performance in terms of COD and BOD_5 removal

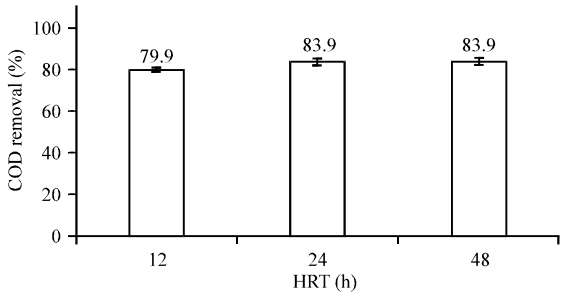

Fig. 3: Effect of HRT on SBR performance in terms of COD removal

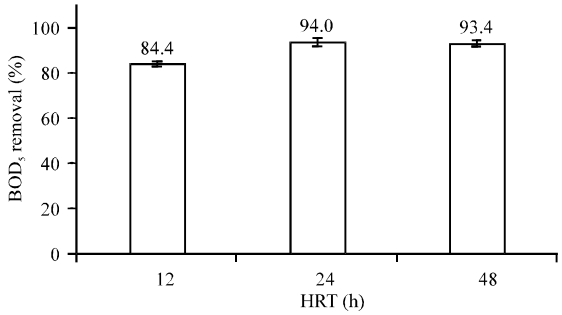

Fig. 4: Effect of HRT on SBR performance in terms of BOD_5 removal

Table 2: Significance of the difference between two means for COD removal in SBR1 and SBR2 under different operating conditions, using Tukey HSD

	MLSS:HRT (mg L^{-1}:h)					
	SBR2			SBR1		
MLSS:HRT (mg L^{-1}:h)	4000:12	4000:24	4000:48	6000:12	6000:24	6000:48
SBR2						
4000:12	1					
4000:24	0	1				
4000:48	0	0.911	1			
SBR1						
6000:12	0.317	0	0	1		
6000:24	0	0.99	0.995	0	1	
6000:48	0	0.371	0.963	0	0.67	1

and is not significantly different if the value of significance is more than 0.05. As it can be seen in Table 2, no remarkable improvement in SBR efficiency in terms of COD removal is seen due to increase of MLSS concentration from low (4000 mg L^{-1}) to high (6000 mg L^{-1}) value. At low MLSS concentration, increasing HRT from 12 to 24 h significantly improved the SBR performance (p-value 0<0.05). However, increasing HRT from 24 to 48 h did not significantly improve the SBR performance (p-value 0.911>0.0). This indicates that most of the substrate degradation occurred during the first 12 h and a smaller portion was degraded in rest of the retention time. This agrees well with the studies reported in the literature for treatment of antibiotic wastewater by combined Fenton-SBR (Elmolla and Chaudhuri, 2012) and combined

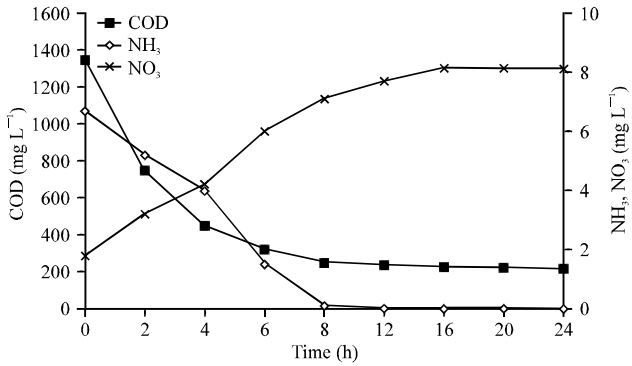

Fig. 5: Biodegradation of the pharmaceutical wastewater in terms of COD, NH_3–N and NO_3^--N in SBR2 during one cycle period under the optimum conditions

photo-Fenton-SBR (Elmolla and Chaudhuri, 2011). Based on the analysis, the optimum operating conditions were MLSS 4000 mg L^{-1} and HRT 24 h.

Performance of SBR under optimum operating conditions: To study biodegradation of the organic carbon and nitrogen under the optimum operating conditions, COD, NH_3-N and NO_3^--N were measured during the optimum cycle period (24 h) for SBR2 (MLSS 4000 mg L^{-1}) and is shown in Figure 5. As shown in the figure, most of the substrate degradation occurred during the first 12 h and a smaller portion was degraded in rest of the retention time. Oxidation of NH_3-N was complete in 8 h and NO_3^--N concentration was 8.2 mg L^{-1} in 16 h, indicating complete nitrification. Under the optimum operating conditions (HRT 24 h and MLSS 4000 mg L^{-1}); the SBR achieved an efficiency of 94±1.3% in terms of BOD_5 removal and 83.9±1.7% in terms of COD removal and complete nitrification occurred in the SBR. The results are comparable with that reported in the literature. Ng *et al.* (1991) reported 90% COD removal by SBR treatment of pharmaceutical wastewater. Also, efficiency of 97% for COD removal was reported for slaughterhouse effluent treatment (Hadjiniculaou, 1998). Ganjidoust and Ayati (2004) reported SBR efficiency of 92, 84, 52% for COD, turbidity and TSS removal, respectively for treatment of wood fiber industry effluent.

CONCLUSIONS

Hydraulic Retention Time (HRT) of 24 h was found suitable for the SBR and increasing HRT from 12 to 24 h significantly improved the SBR performance. Increasing MLSS from 4000 to 6000 mg L^{-1} did not significantly improve the SBR performance for the pharmaceutical wastewater treatment. Under the optimum operating conditions (HRT 24 h and MLSS 4000 mg L^{-1}), the SBR achieved an efficiency of 94±1.3% in terms of BOD_5 removal and 83.9±1.7% in terms of COD removal, with complete nitrification.

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