# Improving Treatment Performance of a Sequencing Batch Biofilm Reactor (SBBR) for Wastewater from a Paper Mill Making Coated Products

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As a great amount of chemicals are employed to carry out the coating process of paper, the wastewater from paper mill making coated products is characterized by high levels of chemical oxygen demand (COD), colour, and total suspended solids (TSS). In this study, wastewater from a paper mill making coated products was treated by a sequencing batch biofilm reactor (SBBR) after a lab-scale coagulation, resulting in COD, colour, and TSS removal efficiencies of 87.7  $\pm$  1.0%, 33.5  $\pm$  5.2%, and 41.4  $\pm$  3.7%, respectively, which exceeded the biological treatment performance in the paper mill. The removal of COD and colour was attributed to the removal of recalcitrant organic matter, and the removal of TSS was attributed to the biofilm.

Keywords: SBBR; Coating wastewater; Paper mill; Biological treatment

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# INTRODUCTION

Coating has been adopted extensively for paper to achieve the desired surface characteristics. The chemicals involved in the coating process, including solvents, surfactants, and mineral salts, are discharged in the wastewater of the paper mill making coated products, which is characterized by strong colour, high chemical oxygen demand (COD), and a large amount of total suspended solids (TSS) (Pokhrel and Viraraghavan 2004).

Generally, biological treatment, which serves to decrease most of the dissolved and colloid organic contaminants (Palumbo *et al.* 2015), can be effective for the wastewater treatment of paper mill, but it cannot guarantee a satisfactory removal for all the pollutants due to recalcitrant compounds that are difficult to degrade (Rodrigues *et al.* 2008) such as lignin and its derivatives (Hubbe *et al.* 2016). Therefore, many attempts have been made on improving the performance of biological treatment in paper mill wastewater treatment. For instance, aerobic granular sludge can achieve better biological treatment performance than conventional flocculent sludge for paper mill wastewater (Horta Morais *et al.* 2016) and cyanobacteria can improve the degradation of dichloroacetate in paper mill effluents, suggesting that conventional biological performance of paper mill wastewater can be improved by various methods (Kirkwood *et al.* 2005).

Given that biofilm reactors could achieve excellent performance in industrial effluents treatments (Di Iaconi *et al.* 2002; Zheng *et al.* 2016), it is supposed that biological treatment performance for coating wastewater can be improved using biofilm reactors. Therefore, in this study, a sequencing batch biofilm reactor (SBBR) was used for treating the wastewater from a paper mill making coated products. The aim of this paper is to evaluate the performance of SBBR for treatment of wastewater from a paper mill making

coated products by comparing it with the performance of biological treatment in the paper mill wastewater treatment system (BTPW).

# **EXPERIMENTAL**

#### **Materials**

The wastewater was taken from a paper mill making coated products in Guangdong Province, southern China. The scheme of the paper mill wastewater treatment system is shown in Fig. 1. The wastewater sent to our lab was treated by screenings and grit and sand removers. The COD, biochemical oxygen demand (BOD), TSS, and colour for the wastewater sent to the lab ranged from 3200 to 3600 mg/L, 970 to 1300 mg/L, 250 to 300 mg/L, and 2600 to 2900 C.U., respectively, with a pH range of 6.9 to 7.3. The sludge from a natural lake located in Guangzhou, Southern China, served as the inoculum for the SBBR.

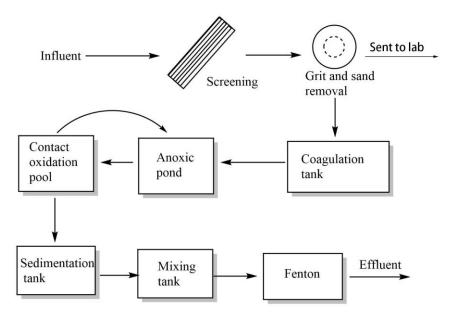


Fig. 1. Scheme of paper mill wastewater treatment system

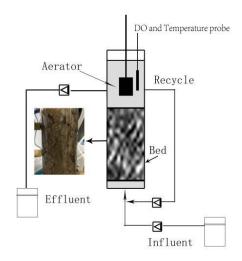
# **Preliminary Treatment**

A lab-scale coagulation was carried out on the wastewater as a preliminary treatment before the biological treatment. The coagulation began with adding PAC (400 mg/L) into the wastewater, and the wastewater was stirred by a magnetic stirring apparatus operating at 240 r/min for 6 min. Cationic polyacrylamide (CPAM) (6 mg/L) was added, and the wastewater was stirred at 100 r/min for 30 min. After 30 min of sedimentation, the coagulation was accomplished with taking the supernatant.

# Lab-scale SBBR

Reactor description and operation

The main bioreactor used in the experiments is a lab-scale SBBR (Fig. 2.) and consisted of a 500 mm high plexiglas cylinder with internal diameter of 60 mm, divided into part A and part B (bed). Plastic packings were filled in part B, and an aerator (ACO-9601, Guangdong Hailea Group Co., Ltd, Chaozhou, China) was placed in part A.



#### Fig. 2. Scheme of SBBR

An operation cycle of the SBBR was 6 h. The operation of the SBBR was divided into filling (10 min), reaction (340 min), and withdrawing (10 min). During the filling phase, a peristaltic pump (BT100-1J, Baoding Longer Precision Pump Co., Ltd, Baoding, China) was used to supply the influent. During the reaction phase, a peristaltic pump (7593-07, Cole-Parmer, Vernon Hills, USA) was used to recycle the wastewater, with a flow rate of 30 L/h. The reaction stage was further divided into an anaerobic phase and an aeration phase by controlling the aerator. When the aerator was turned on, it allowed the DO of the reactor to be kept at 4.5 to 6.0 mg/L. The effluent was removed from a port over a motorized valve during the withdrawing. The effective volume of the SBBR was 900 mL, and 300 mL seed sludge was added into the reactor in advance.

The start-up of the SBBR is shown in the Appendix. After start-up, the SBBR was operated under condition of 9 h of HRT, 1 h of anaerobic time and 5 h of aeration time for 90 days.

#### Analytical methods

COD, BOD, TSS, and colour were measured according to standard methods (APHA 2005). A pH meter (PB-10, Sartorius, Gottingen, Germany) was adopted to monitor pH. A DO meter (Sension 6, Hach, Loveland, CO, USA) was used for measuring DO.

#### **RESULTS AND DISCUSSION**

The characteristics of SBBR effluent and paper mill secondary effluent, as well as the coagulation effluent (influent), are summarized in Table 1, in terms of composition range. The treatment performance of SBBR on COD, colour, and TSS is shown in Fig. 3.

		3			
Item	Unit	Paper Mill Secondary Effluent	Coagulation Effluent	SBBR Effluent	
pН	-	6.4 - 7.8	6.9 - 7.3	7.2 - 7.6	
COD	mg/L	180 - 220	920 - 960	96 - 133	
BOD	mg/L	-	460 - 630	0.5 - 8.8	
Colour	C.U.	20 - 30	36 - 44	23 - 28	
TSS	mg/L	6 - 10	8.4 - 10.6	4.7 - 6.2	

 Table 1. Composition Range of Different Effluents

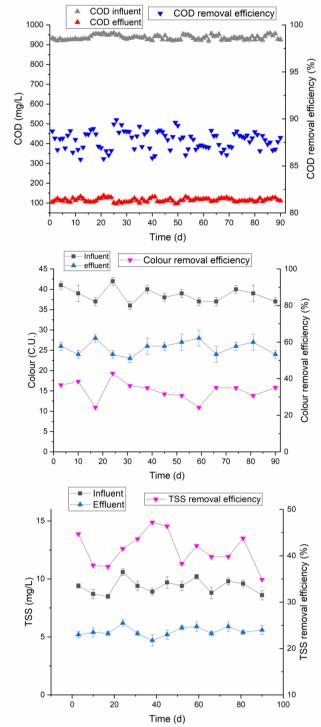


Fig. 3. Treatment performance of SBBR: (a) COD, (b) Colour, (c) TSS

The concentrations of COD, colour, and TSS of the influent (coagulation effluent) were 936.1  $\pm$  14.4 mg/L, 38.6  $\pm$  1.8 C.U., and 9.4  $\pm$  0.6 mg/L respectively; of SBBR effluent were 115.0  $\pm$  9.2 mg/L, 25.6  $\pm$  1.7 C.U., and 5.5  $\pm$  0.4 mg/L respectively. SBBR achieved COD, color and TSS removal efficiencies of 87.7  $\pm$  1.0%, 33.5  $\pm$  5.2%, and 41.4  $\pm$  3.7% respectively. The results suggested that SBBR achieved a better treatment performance than BTPW, especially on COD and TSS.

The SBBR was operated intermittently through anaerobic and aerobic operations in an individual reactor, which provided a balance between anaerobic and aerobic microorganisms that enabled the removal of COD to be achieved. Previous studies indicated that some recalcitrant compounds could be removed by anaerobic digestion (Xiang et al. 2016) and aerobic digestion is effective for treatment of paper mill effluent (Vashi et al. 2018). Therefore, the COD removal could be attributed to the efficient removal of biodegradable compounds; however, recalcitrant compounds were also partly removed by the SBBR, resulting in a COD removal higher than the BTPW. This result confirms that of Pathiraja et al. (2019), who found that polychlorinated biphenyls could be effectively degraded using alternating anaerobic aerobic treatments. Cai et al. (2019) showed that recalcitrant compounds in the secondary effluent of paper mill can be removed by a SBBR. Osman et al. (2013) used a granular activated carbon sequencing batch biofilm reactor to treat recycled paper mill wastewater and achieved a high COD removal. Kuang et al. (2018) found that lignin and its derivatives, the most representative recalcitrant compounds in paper mill effluents, can be effectively degraded by a SBBR. In this study, SBBR enhanced the removal of COD by degrading refractory substances in the wastewater such as lignin and its derivatives.

The color of wastewater, on which the conventional biological treatment system has little effect, is one of the main environmental problems in the pulp and paper industry. According to Muhamad *et al.* (2015), the colour of paper mill effluent is generally formed by the presence of lignin and its derivatives, which are difficult to be degraded naturally. The color of wastewater was lowered by SBBR due to the partial degradation of refractory organic matter such as lignin and its derivatives, which was consistent with the COD removal performance. Similarly, Lotito *et al.* (2014) observed effective color removal of textile wastewater, which was characterized by high content of recalcitrant compounds, during a treatment using SBBR. Thus, SBBR removed recalcitrant compounds better than conventional bioreactors so it achieved better removal effects on COD and colour.

Although coagulation resulted in efficient removal of TSS, SBBR was able to reduce it further. Conventional bioreactors that depend on gravity separation for the removal of TSS are greatly influenced by settling time (Yong *et al.* 2018). The reduced TSS in SBBR can be ascribed to biofilm, which absorbs insoluble pollutants in the wastewater (Rittmann 2018). As biofilm reactors achieve TSS removal as high as 99.9% (Di Iaconi *et al.* 2002), SBBR adsorption is a better alternative than gravity separation to remove TSS. Therefore, it is speculated that using SBBR could improve biological treatment performance by removing soluble recalcitrant compounds and insoluble contaminants.

# CONCLUSIONS

1. A sequencing batch biofilm reactor (SBBR) achieved chemical oxygen demand (COD) and colour removal efficiencies of  $87.7 \pm 1.0\%$ ,  $33.5 \pm 5.2\%$ , which has exceeded the performance of the biological treatment in the paper mill wastewater treatment system (BTPW). In the SBBR, COD and colour removal can be attributed to the removal of recalcitrant organic matter.

2. SBBR achieved a total suspended solids (TSS) removal efficiency of  $41.4 \pm 3.7\%$ . The TSS removal can be ascribed to the biofilm, which absorbs insoluble pollutants in the wastewater.

# ACKNOWLEDGMENTS

This research was supported by the Natural Science Foundation of Guangdong Province (2014A030310145).

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Article submitted: April 19, 2019; Peer review completed: July 20, 2019; Revised version received: August 4, 2019; Accepted: August 6, 2019; Published: August 16, 2019; DOI: 10.15376/biores.14.4.7992-8001

# APPENDIX

#### METHODS

After sludge was added into the SBBR, sludge cultivation and acclimation were carried out successively, followed by detecting the optimal HRT and aeration time in a cycle. The methods and results during these periods are shown in Supplementary material A. The SBBR was operated under condition of 9 h of HRT, 1 h of anaerobic time, and 5 h of aeration time for 90 days.

# Sludge Cultivation and Acclimation

Nutrient solution made of glucose, ammonium nitrate and dipotassium phosphate according to a certain C:N:P (100:5:1) ratio was supplied to cultivate the sludge. The COD of the influent ranged from 400 mg/L to 1400 mg/L. The acclimation of sludge was carried out after the cultivation stage of the sludge as nutrient solution (COD of 200 mg/L) and coagulated wastewater were mixed according to certain proportions (3:1,1:1 and 1:3) as influent and supplied to the reactor.

During the cultivation and acclimation stage, the HRT was set as 24 h. While an operation cycle was set as 6 h, the first 3 h belonged to anaerobic phase as the aerator was switched off. The next 3 h belonged to the aeration phase as the aerator was turned on.

# **Aeration Time and HRT**

After the acclimation stage, the coagulated wastewater was served as the influent and the reactor was operated under condition of 24 h HRT, 3 h anaerobic time, and 3 h aeration time for 12 d. After then, the SBBR was operated under different HRT and different aeration time. The operation of SBBR was therefore divided into 6 phases according to HRT and aeration time, with each phase lasting for 10 d. The information of each phase was listed in table 1.

Items	Phase						
	1	2	3	4	5	6	
Days	10	10	10	10	10	10	
HRT (h)	18	18	12	12	9	9	
Anaerobic time (h)	4	3	2	1	1	0	
Aeration time (h)	2	3	4	5	5	6	

# Table S1. Different Phases of the SBBR

# RESULTS

# Sludge Cultivation and Acclimation

The COD removal of SBBR during the cultivation period is presented in Fig. 1. With the supply of the nutrient solution, the removal of COD increased from about 50% to 95% during the 15 cycles of cultivation stage. The COD of influent also increased from 800 to 1400 mg/L during the same time. Due to that 1400 mg/L has already surpassed the COD range of the coagulated wastewater, the sludge was proposed to have satisfied the requirements of acclimation.

The sludge acclimation was implemented after the cultivation stage, with the COD range and removal during this period being presented in Fig. 2. In general, a tendency

of firstly going up and then stabilizing of COD removal was presented due to the fact that the activated microorganism may require some time to adapt to the pulping wastewater. 8 cycles and 6 cycles were taken for the COD removal to increase from 40% to 60% and from 50% to 60%, with the the COD of effluent ranging from 300 mg/L to 400 mg/L and 600 to 700 mg/L, respectively. After that, a stabilized COD removal of around 60% was rapidly reached when the coagulation effluent was directly added into the SBBR. Eventually, with the sludge being adopted by the packings in the biofilm area and the wastewater in the aeration area being clarified, the sludge acclimation was proposed to be accomplished.

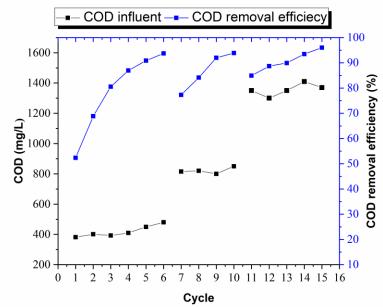


Fig. S1. Treatment results of SBBR reactor during sludge cultivation stage

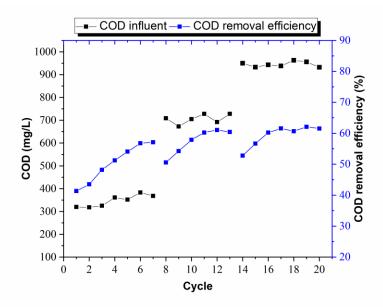


Fig. S2. Treatment results of SBBR reactor during sludge acclimation stage

# Aeration time and HRT

The SBBR performance under different HRT and aeration time was detected and shown in Fig. 3. With the anaerobic reaction time being 4 h, aeration time being 2 h and HRT being 18 h, the COD was reduced from roughly 922.7 mg/L to around 448.6 mg/L, indicating a COD removal of 51.4%. Considering that the anaerobic reaction time was longer than the aerobic reaction time, the anaerobic bacteria was supposed to be the dominant microorganism and may play a major role in digesting the pollutants.

When the aeration time was increased and the anaerobic reaction time was decreased, the COD removal experienced great changes. Firstly, the COD removal increased to 75.6% as the COD was decreased from around 941.7 mg/L to around 226.6 mg/L, with the anaerobic reaction time being 2 h, the aeration time being 4 h and the HRT being 12 h. Then, the COD removal surpassed 60% with aeration time being 2 h, anaerobic time being 4 h under the same HRT. Eventually, when the HRT was reduced to 9 h, the COD removal efficiency reached 87.6% with 1 h of anaerobic reaction time and 5 h of aeration time and 87.1% with 6 h of aeration time.

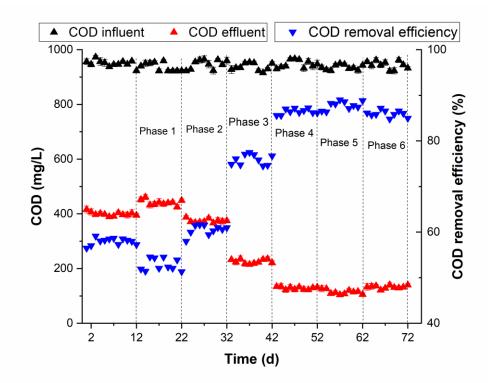


Fig. S3. COD removal of SBBR under different HRT and aeration time