Application of Granular Activated Carbon-Sequencing Batch Reactor (GAC-SBR) System for Treating Pulp and Paper Industry Wastewater

Suntud Sirianuntapiboon

Deaprtment of Environemntal Technology, School of Energy and Materials, king Mongkut's University Thonburi, Bangkok 10140, Thailand.

Abstract

The study was concerned with removal of organic matters (COD, BOD5 and SS) and color substances from pulp and paper industry wastewater by GAC-SBR system. GAC showed the COD and color adsorption yields under jar test conditions as 127.00 mg/g of GAC and 248.00 Pt-Co/g of GAC, respectively. Under SBR conditions (full aeration), the COD and color removal abilities of GAC could increase about 3.16% and 1.05%, respectively. The COD and color removal abilities of GAC were decreased after use in the SBR system. The COD and color removal abilities of 10 daysused GAC and 20 days-used GAC were reduced to 49.02 mg/g GAC and 217.09 Pt-Co units/g GAC and 58.82 mg/g GAC and 205.68 Pt-Co units/g GAC respectively. But, after 30 days operation of GAC-SBR system, the COD adsorption ability of GAC was increased to 107.85 mg/g GAC due to the biological regeneration of GAC by bio-sludge. In the SBR system, the impurities of wastewater such as COD, BOD₅ could be removed with high efficiency but the color removal efficiency was quite low. The COD, BOD₅ and color removal efficiencies of SBR system were about 73.26%, 95.10 and 56.96%, respectively under HRT 1 day. However, when the HRT of the system was increased, the COD and BOD₅ removal efficiencies were increased and the color removal efficiency was decreased. The COD, BOD₅ and color removal efficiencies of the system under HRT of 10 days were increased to 89.80% and 97.84%, while the color removal efficiency was decreased to 47.63%. GAC in the GAC-SBR system could increase the ability of the system for reduction of both COD and color substances. The COD, BOD₅ and color removal efficiencies of the system under HRT of 10 days were up to 90.60%, 97.84% and 52.94%, respectively.

Keywords: Granular activated carbon (GAC)/ Sequencing batch reactor (SBR)/ Granular activated carbon sequencing batch reactor (GAC-SBR) / Pulp and paper industry wastewater / Adsorption.

Offprint requests to Dr. Suntud Sirianuntapiboon.

1. Introduction

Wastewater problems have become a serious problem in the world. The volumes of wastewater from domestic and industrial sectors are increasing year by year. Pulp and paper industry is one agroindustry that uses large amounts of water for washing, boiling. The wastewater from pulp and paper contains not only high COD concentration but also high color intensity [1]. The quality of wastewater is different in each factory due to the raw materials and production processing. The wastewater

treatment systems, used in the pulp and paper industry, are biological treatment process such as activated sludge system, oxidation pond, combined system of activated sludge and oxidation pond and combined system of anaerobic pond and oxidation pond [1,2]. But activated sludge system is one of selected system due to its high removal efficiency, suitable for this type of wastewater. But, several problems would occur during operation such as sedimentation tank and volume of excess sludge [2,3]. To solve the above problems, several researchers tried to use the sequencing batch

reactor system (SBR). Then, SBR system was introduced for treating the pulp and paper industry wastewater [4,5,6,7]. Franta et al [6] reported that COD removal efficiency was increased with the increasing of aeration period. The removal efficiency of the SBR system depended on the sludge age [7]. However, by using the SBR system, color substances in the wastewater still remained. Additionally, it is well known that activated carbon is a good adsorbent [8,9,10] for adsorbing of color, odor suspended solids and so on. Actually, activated carbon is wildly used in water purification, wastewater treatment, air purification and air pollution treatments by physical adsorption mechanism [8.9]. Furthermore, activated carbon could be used together with the biological treatment processes to increase the removal efficiency and reduce HRT in biological treatment system. Many experiments were carried out for comparing of BOD₅ removal efficiency by using activated sludge system with without supplementation of powder activated carbon (PAC) [11,12,13,14,15,16]. The results showed that PAC could reduce HRT of the system and increase the removal efficiency [17,18,19]. But many problems were occurred during operation of activated sludge system such as the contamination of PAC in effluent, the loss of PAC with discharging of excess sludge and the increasing sedimentation time [17].

In this study, we have tried to introduce the sequencing batch reactor system with adding granular activated carbon (It is called granular activated carbon sequencing batch reactor system (GAC-SBR) for treating pulp and paper industry wastewater. The optimum COD and color adsorption abilities of GAC were investigated. The optimal HRTof both SBR and GAC-SBR systems were also investigated for observation of the highest removal efficiency.

2. Materials and Methods

Pulp and paper industry wastewater (PPWW): The wastewater of the wastewater treatment of Siam Kraft Pulp and Paper Industries Co., Ltd, Ban-Prong, Rajchaburi, Thailand was used in this experiment as the PPWW.

Granular Activated Carbon (GAC): The GAC which was used in this experiment was CGC-11 with a mesh size of 8x30 mm². The

specifications of the GAC were a total surface area of 1050-1150 m²/g and apparent density of 0.46-0.48 g/ml, respectively.

Sequencing Batch Reactor System (SBR): Six 10-liter reactors, made from acrylic plastic (5 mm thick) were used in this experiment as shown in fig.1. The dimensions of the reactor were 18 cm in diameter and 40 cm in height. The working volume was 7.5 litters. Low speed gear motors, model P 630A-387, 100V, 50/60 Hz, 1.7/1.3 A (Japan Servo Co. Ltd., Japan) were used for driving the paddle shape impeller. The speed of the impeller was adjusted to 60 rpm. One air-pump system, model EK-8000, 6.0 W (President Co. Ltd., Thailand), was used for supplying air for 2 sets of reactors.

Acclimatization of bio-sludge for SBR system: Sludge from the wastewater treatment plant (activated sludge system) of Siam Craft Pulp and paper Industries Co., Ltd was used as an inoculum of SBR and GAC-SBR systems. The sludge was cultivated in PPWW for 1 week before being used in the experiments.

Chemical adsorption ability test of GAC: The PPWW of Siam Craft Pulp and paper Industries Co., Ltd was used in this experiment for testing the adsorption capacity of GAC under jar test conditions and SBR conditions (aeration condition). The adsorption capacity of GAC on and color substances, at various concentrations of GAC (500, 1,000, 1,500, 2,000 and 2,500 mg/l) were collected and analyzed by using Freundlich's adsorption isotherm equation [5] for the determination of maximal adsorption capacity of GAC for COD and color intensity. The used GAC was also investigated for determination of color and COD adsorption capacity. The used GAC which collected from SBR system after being operated for 10, 20, and 30 days was collected for use as 10 days-used GAC, 20 days-used GAC and 30 days-used GAC, respectively.

Operation of SBR and GAC-SBR system: For the SBR system, 1.4 liters of 10,000 mg/l of acclimatized sludge solution was inoculated in each reactor (the final concentration of sludge in each reactor was 2,000 mg/l). And then, PPWW was added up to the maximum capacity of 7,500 ml within 1 hr. During feeding of PPWW, the system had to be aerated fully and then continuously aerated for 19 hrs. Aeration was then shut down for 3 hrs. After the sludge had fully settled, the

supernatant had to be removed (about 7,000 ml) within 0.5 hr and the system must be kept under anoxic condition for 0.5 hrs. After that the new PPWW solution (about 7,000 ml) was filled into the reactor and the above operation was repeated. For the GAC-SBR system, the operation conditions were similar to SBR system, but the 7,500 mg of GAC had to be added in each reactor (the final concentration of the GAC in the system was 1,000 mg/l).

Determination of removal efficiencies for SBR and GAC-SBR systems in various HRT: The experiments were carried out in ten liters reactor, with and without GAC supplemented at a final concentration of 1000 mg/l under various HRT condition as1, 3, 5 and 10 days. The sludge age (solid retention time: SRT) was controlled at 10 days. The effluents of the system were collected for determination of chemical properties.

Chemical Analysis: The COD, BOD₅, mixed-liquor suspended solids (MLSS), suspended solids (SS), pH and color intensity of influents and effluents were determined by using standard methods for the examination of water and wastewater [20].

3. Results

The Chemical Properties of Pulp and Paper Industry wastewater: The wastewater samples from the sump tank of wastewater treatment plant in Siam Kraft Pulp and Paper Industries Co., Ltd, Ban-Prong, Rajchaburi, Thailand was collected for chemical properties analysis. The chemical properties of this wastewater are shown in table1. The COD concentration in the wastewater fluctuated. It varied from 979 mg/l to 1712 mg/l. The COD: BOD₅ ratio was 2.8:1. The color intensity of wastewater was in the range of 547-764 Pt-Co units.

Optimum adsorption capacity of GAC for COD and color substances: The results are shown in table2. The GAC shows the high adsorption ability in both organic matters (COD) and color substances under jar test conditions as 127.00 mg/g GAC and 280.00 Pt-Co units/g GAC, respectively as shown in table2. However, the aeration (SBR system condition) could increase both COD and color adsorption capacities of GAC. The COD and color adsorption abilities of GAC were increased to 131.01 mg/g GAC and 250.62 Pt-Co units/g

GAC, respectively. It meant that the COD and color adsorption abilities of GAC increased 1.05%, respectively. 3.16% and about Concerning the adsorption ability test of used GAC, the results are shown in table2. The COD adsorption ability of GAC decreased from 131.01 mg/g GAC to 49.02 mg/g GAC when the GAC was used in the GAC-SBR system for 10 days. But, the COD adsorption ability of GAC recovered when it was cultivated in GAC-SBR system for more than 10 days. The COD adsorption ability of 20 days-used GAC and 30 days-used GAC were 58.82 mg/g GAC, and 107.85 mg/g GAC, respectively. The Color adsorption ability of GAC decreased with the increasing of the using period of GAC in the GAC-SBR system as shown in table2. The color adsorption ability of 30 days used GAC reduced from 250.62 Pt-Co units/g GAC to 174.96 Pt-Co units/g GAC when compared with the new GAC.

Effects of GAC on the removal efficiency of SBR system: Under the SBR condition, the impurities of the wastewater could be removed as shown in table3 and fig.2, fig.3, fig.4 and fig.5. The results show that the COD, BOD, SS and color intensity of effluent from the SBR system were quite stable during operation for more than 4 weeks as shown in fig.2, fig.3, fig.4 and fig.5. The average COD, BOD, SS concentration and color intensity of effluent from the SBR system under HRT of 1 day were 140 mg/l, 22.0 mg/l, 17.0 mg/l and 300 Pt-Co units, respectively as shown in table3. This means that the COD, BOD, color and SS removal efficiencies of the SBR system were about 73.26%, 95.10%, 56.96% and 90.05%, respectively. When the GAC was added in the SBR system, the removal efficiencies of the system were increased as shown in table3 and fig.2, fig.3 fig.4 and fig.5. The COD, BOD, SS and color intensity of effluent from GAC-SBR system under HRT of 1 day were 128 mg/l, 20 and 290 Pt-Co units, mg/l respectively as shown in table3. Therefore, the COD, BOD, color and SS removal efficiencies of GAC-SBR system were increased to 58.39% and 92.10%, 89.806%, 95.99%, respectively.

Effect of HRT on the removal efficiencies of SBR and GAC-SBR systems: The removal efficiencies of both SBR and GAC-SBR on the COD, BOD and SS rose with an increase of

HRT, as shown in table3, fig.2, fig.3, fig.4 and fig.5. In the SBR system, the COD, BOD and SS removal efficiencies increased from 73.26%, 95.10% and 90.05% to 90.83%, 95.79% and 92.425% and 90.76%, 97.84% and 93.00% respectively when the HRT of the system was increased from 1 day to 3 days and 5 days respectively as shown in table3. The COD, BOD and SS concentration of the effluent from the system in each day was not so much different during operation for more than 4 weeks as shown in fig.2, fig.3 and fig.4. For the experiments in the GAC-SBR system, it showed the same patterns of removal efficiencies as in the SBR system, but the removal efficiencies of the GAC-SBR system were higher than in the SBR system under the same HRT operation as shown in table3. The COD, BOD and SS removal efficiencies increased from 89.80%, 95.99% and 92.10% to 91.31%, 98.60% and 95.42% and 91.47%, 97.94% and 94.26% respectively when the HRT of the system was increased from 1 day to 3 days and 5 days respectively. For the color removal efficiency of both the SBR and the GAC-SBR systems, the results show that the removal efficiency decreased with an increase of HRT, as shown in table3. For example, the color removal efficiencies of the SBR and the GAC-SBR systems deceased from 56,96% and 58.39% to 47.63% and 52.945%, respectively.

Effects of GAC and HRT on the quality of the sludge in SBR system: The results are shown in table4. The sludge volume index (SVI) of the system decreased with an increase of HRT value. For example, the SVI of the SBR system under HRT of 1 day was 120 ml/g of sludge, but it decreased to 100 mg/g of sludge, 80 ml/g of sludge when the HRT of the system from 80 to 90 when the HRT of the system was up from 5 days to 10 days as shown in table4. For the effects of GAC on the quality of sludge of SBR system, the results in the table4 show that the GAC could increase the quality of the sludge. The SVI of sludge under HRT of 1 day decreased from 120 ml/g of sludge to 80 ml/g of sludge when the GAC was added into the system at final concentration of GAC of 1,000 mg/l.

4. Discussions and Conclusions

Wastewater from pulp and paper industry is one of the serious problems due to the high

concentration in COD and color substances [1]. Several treatment systems heve been introduced for treating this kind of wastewater on the basis of its high removal efficiency and low operating cost [2]. Siam Kraft pulp and paper Industries Co., Ltd, one of the biggest companies to produce pulp and paper products in Thailand, was used for checking the wastewater quality. The wastewater from this factory had high concentration of COD, BOD5 and color substances as 1255 mg/l, 449 mg/l and 697 Pt-Co units, respectively. The COD: BOD₅ ratio was 2.8:1. This means that the pulp and paper industry wastewater contains large amounts of organic compounds that are difficult to biodegrade and inorganic reducing substances [2,6,15,20]. However, most of the pulp and paper industry factory still uses the biological treatment system such as the oxidation pond, aerated lagoon or activated sludge [8,9]. Many factories have problems with the fluctuation in the quality of the effluent, stability of the system and, operating and investment cost. The SBR system has been introduced into several kinds of industries such as seafood and meat processing industries for reduction of investment and operating costs [4,5,6]. In this study, we have tried to introduce the SBR system to treat pulp and paper industry wastewater [6]. The results show that the microorganisms in SBR system can remove both organic matters (COD and color substances from and wastewater as shown in table3. The effluent qualities, except color intensity passed the standard permission of Department of Industrial Works of Thailand (21) when the SBR system was operated under HRT of 3 days. And also, the removal efficiencies of SBR system improved with the increase of HRT, except for color removal efficiency as shown in table3. The COD removal efficiencies of the SBR system were increased from 73.26% to 89.80% while the color removal efficiency was reduced from 56.96% to 48.92% when HRT of the system was increased from 1 day to 3 days. It means that when the HRT was increased, the organic loading was decreased then; the removal efficiency was increased [2,3,4]. But the color removal efficiency was decreased with the increase of HRT due to the decrease of excess sludge of the system, because the color substances in the wastewater were removed by both bio-adsorption into the cell (bio-sludge)

and physical adsorption into GAC, as shown in table2. Then, the color removal yield was increased mainly with increase of the bio-sludge concentration in both GAC-SBR system and SBR system, [8,9,10]. However, the quality of the sludge (SVI) in SBR system was deceased with the increase of HRT as shown in table4. Then, the GAC added into the SBR system to increase the quality of sludge and removal efficiencies and reduction of the HRT [8,9,11]. GAC, itself, could adsorb both COD and color substances from the wastewater, as shown in table1. The adsorption ability of GAC could be increased by aeration, because of the increase in the mixing efficiency and adsorption ability of GAC [8]. The GAC also could reduce the value of SVI. So, It can be said that both SBR and GAC-SBR system could apply to treat the wastewater from the pulp and paper industry. For the SBR system, the organic matters in the wastewater were easier to remove but the color substances were removed by bio-adsorption mechanisms. Then the color removal efficiency of SBR system could be increased with a decrease of the sludge-age or increase of the excess sludge volume. However, by increase the amount of sludge, the sludge-age was reduced, thereby improving its adsorbative qualities. Then, the GAC was used to inprove the sludge quality (reduction of SVI value) And also the HRT of SBR system could reduce by adding GAC in to the system. For example, the GAC-SBR system under HRT of 1 day and SBR system under HRT of 3 days gave almost the same removal efficiency. And the GAC-SBR system gave more advantage on the color removal efficiency as shown in table3.

For the application, it could be said that both SBR and GAC-SBR systems could be used for treating the wastewater from the pulp and paper industry. But, the GAC-SBR system might be most suitable and the best system due to the effects of GAC on the physical adsorption ability and its ability to improve the quality of sludge. And the adsorption ability of GAC was recovered by biological regeneration during the operation under GAC-SBR system [22].

5. Acknowledgements

We wish to express our thanks to Department of Environmental Technology, School of Energy and Materials, KMUTT and Siam Kraft Pulp and Paper Co., Ltd. for providing the research materials and funds. Also, the deep thanks is given to the Chin Sophonpanith foundation for supporting funds. Finally, sincere thanks is given to Miss Laddawan Rattanaprathipthong for her technical assistance.

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<u>Table1</u>: Chemical properties of wastewater of Siam Craft pulp and paper Co., Ltd.

Parameters	Value				
	Range	Average			
COD (mg/l)	979-1712	1255			
BOD (mg/l)	344-623	449			
pH	4.95-6.78	6.06			
SS (mg/l)	74-264	190			
Color intensity (Pt-Co unit)	547-764	697			

<u>Table2</u>: Adsorption capacity of GAC on COD and color substances.

Type of GAC	Adsorption capacity						
31	Under ja	ar test condition	Under SBR system condition				
	COD	Color substances	COD	Color substances			
	(mg/g GAC)	(Pt-Co unit/g GAC)	(mg/g GAC)	(Pt-Co unit/g GAC)			
New GAC	127.00	248.00	131.01	250.62			
10 days-used GAC	-	-	49.02	217.09			
20 days-used GAC	-	-	58.82	205.68			
30 days-used GAC	_	-	107.85	174.96			

Table3: Effects of HRT on efficiencies of SBR and GAC-SBR systems.

Parameter			HRT of the system							
			1 day		3 days		5 days		10 days	
	Influent		SBR	GAC-SBR	SBR	GAC-SBR	SBR	GAC-SBR	SBR	GAC-SBR
COD	1,255	Effluent (mg/l)	140.0	128.0	115.0	109.0	116.0	107.0	128.0	118.0
		% Removal	73.26	89.80	90.83	91.31	90.76	91.47	89.80	90.60
BOD	449	Effluent (mg/l)	22.0	20.0	10.5	9.7	10.8	10.3	11.5	10.8
		% Removal	95.10	95.99	95.79	98.60	97.84	97.94	97.70	97.84
Color	697	Effluent (mg/l)	300	290	356	347	370	339	365	328
intensity		% Removal	56.96	58.39	48.92	50.22	46.92	51.36	47.63	52.94
SS	190	Effluent (mg/l)	17.0	15.0	14.4	8.7	13.3	10.9	21.5	20.4
		% Removal	90.05	92.10	92.42	95.42	93.00	94.26	88.68	89.26
рН	6.06	Effluent (mg/l)	7.8-8.4	8.0-8.3	8.1-8.7	7.9-8.7	8.4-8.7	8.4-8.7	8.7-9.0	8.6-8.7

Table4: Effects of HRT on sludge volume index of SBR and GAC-SBR system.

Parameter	HRT of the system							
	1 day		3 days		5 days		10 days	
Type of the system	SBR	GAC-SBR	SBR	GAC-SBR	SBR	GAC-SBR	SBR	GAC-SBR
SVI (ml/g of sludge)	120	80	100	75	80	75	90	90

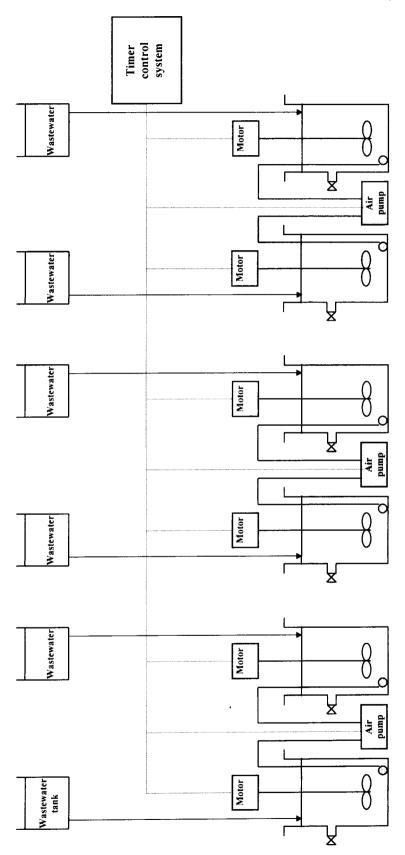


Fig. 1: Flow diagram of SBR treatment system.

pump was supplied for 2 sets of reactor the working volume of the reactor was 75 % of total volume (7.5 litres). The chemical and biological The physical operation controlling were 60 rpm of impeller speed, fully aeration with air-pump system model EK-8000, 6.0 W (one set of air operation controlling were described in the text due to each experiment

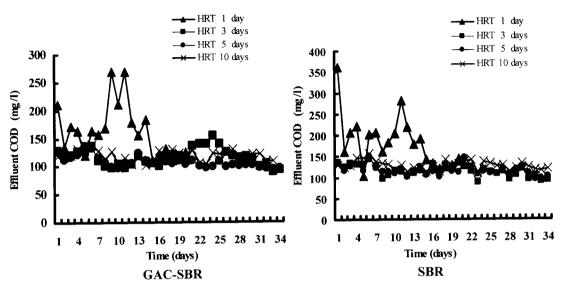


Fig.2: Effluent COD profiles in SBR and GAC-SBR systems in various conditions of HRT. The experiment conditions are described in the text.

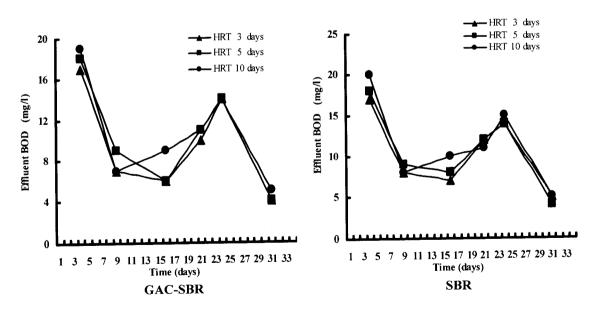


Fig.3: Effluent BOD profiles in SBR and GAC-SBR systems in various conditions of HRT. The experiment conditions are described in the text.

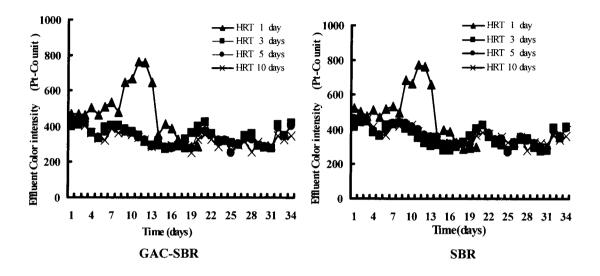
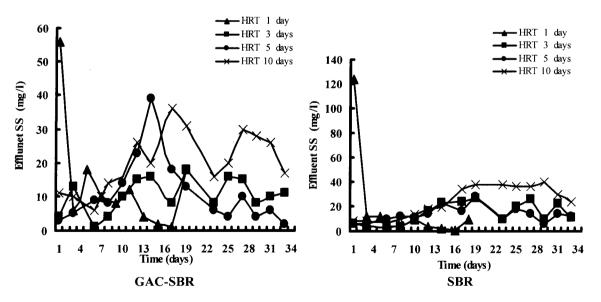


Fig.4: Effluent color intensity profiles in SBR and GAC-SBR systems in various conditions of HRT.

The experiment conditions are described in the text.



<u>Fig.5</u>: Effluent SS profiles in SBR and GAC-SBR systems in various conditions of HRT. The experiment conditions are described in the text.