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Simultaneous removal of AOX and COD from real recycled paper wastewater using GAC-SBBR

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ABSTRACT

A lab-scale granular activated carbon sequencing batch biofilm reactor (GAC-SBBR), a combined adsorption and biological process, was developed to treat real wastewater from a recycled paper mill. In this study, one-consortia of mixed culture (4000–5000 mg/L) originating from recycled paper mill activated sludge from Kajang, Malaysia was acclimatized. The GAC-SBBR was fed with real wastewater taken from the same recycled paper mill, which had a high concentration of chemical oxygen demand (COD) and adsorbable organic halides (AOX). The operational duration of the GAC-SBBR was adjusted from 48 h to 24, 12 and finally 8 h to evaluate the effect of the hydraulic retention time (HRT) on the simultaneous removal of COD and AOX. The COD and AOX removals were in the range of 53–92% and 26–99%, respectively. From this study, it was observed that the longest HRT (48 h) yielded a high removal of COD and AOX, at 92% and 99%, respectively.

1. Introduction

The main source of wastewater containing AOX compounds is the paper mill industry, the oil refining and petrochemical industries, leather manufacturing, textile factories, plastics factories, pesticides factories and pharmaceutical industries (Annachhatre and Gheewala, 1996; Anirudhan et al., 2009). The paper mill industry is the sixth largest polluter discharging a variety of hazardous materials, including AOX, into the environment (Ali and Sreekrishnan, 2001).

AOX is the parameter that serves to assess the total amount of organically bound chlorine and can be expressed as micrograms per liter (μg/L) of chloride ions in wastewater (Bernt et al., 1998). The discharge limit of AOX for a paper mill is in the range of 1–100 μg/L (Latorre et al., 2005). Harrison (2002) maintains that certain countries, such as Sweden, Australia and the US have established AOX targets that are in the range of 0.3–0.6 kg/ADt for the pulp and paper mill industries. AOX, especially chlorophenols from monochlorophenol (MonoCP) to pentachlorophenol (PCP) are widely used as pesticides and wood preservatives (Ladislao and Galil, 2004). These compounds have been identified as harmful to the ecosystem and can act as inhibitors of microorganism growth, with effects ranging from mutagenic to carcinogenic (Pang et al., 2007; Singh et al., 2007a; Hameed et al., 2009).

A single approach involving a biological, chemical or physical treatment is usually not effective enough to meet the increasingly stringent discharge requirements for the pulp and paper mill industries (Thompson et al., 2001). From the literature, there are many types of AOX treatments, such as adsorption (Hameed et al., 2009), biological (Farrell and Quilty, 2002), ozonation (Balcioglu et al., 2007) and UV/photocatalysis (Ou et al., 2006). The adsorption treatment has been chosen by many researchers due to its low cost of operation and simplicity of design compared with other treatments. This treatment requires using an activated carbon which can be generated from various types of resources (Hameed et al., 2009), including those originating from agricultural waste, such as coconut shells, coconut husks, apricot shells, walnut shells and bamboo (Anirudhan et al., 2009). The high adsorption capacities of activated carbons are usually related to their high-surface area, pore volume, and porosity (Ahmaruzzaman, 2008). In addition, activated carbon, which has a coarse texture and a large surface area is suitable for biofilm attachment.

AOX can also be partially or completely degraded by several species of single bacteria (Perez et al., 1997; Goswami et al., 2002; Singh et al., 2007b), mixed culture (Farrell and Quilty, 2002;

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Atuanya and Chakrabarti, 2004), acclimated biomass (Ladislao and Galil, 2004) and biofilms (Carvalho et al., 2001). Biomass or biofilm consist of a mixed culture of microorganisms attached to a coarse surface. These microorganisms are responsible for biodegrading and removing the organics and nutrients in wastewater (Metcalf and Eddy. Inc, 2004). Because AOX is highly toxic for microorganism growth, a combination of adsorption and biofilm processes is an effective treatment process for AOX (Carvalho et al., 2001).

To obtain optimum operation of the wastewater treatment process, the most important parameter that must be considered is the HRT. The effect of the HRT has been investigated in many studies of wastewater treatment using either real or synthetic wastewater. A wide range of HRTs have also been tested in these studies. In most cases, the removal of COD can be as high as 60–95% (Diez et al., 2002; Wang et al., 2007), and the removal of organic compounds is approximately 33–94% (Diez et al., 2002; Krumins et al., 2002; Wang et al., 2007).

Few studies involving real wastewater, such as Kraft mill wastewater, palm oil mill effluent (POME) and leachate have been performed for longer HRTs. In a study by Diez et al. (2002) using an activated sludge treatment for Kraft mill wastewater, the HRT was varied from 4.5 to 48 h for phenolic compounds and nutrient removal and for COD, BOD_5 and TSS removal was from 6 to 16 h. For the POME treatment by Badiei et al. (2011), the HRT ranges from 36 to 96 h, and for the leachate treatment (Krumins et al., 2002), the HRT ranges from 3 to 24 h. Compared with the small-scale experiment using synthetic wastewater, the time used tends to be shorter than 12 h of operation (Kargi and Uygur, 2004; Wang et al., 2007).

According to Muhamad et al. (2011), the main efforts within the pulp and paper industry to control environmental emissions have focused on the control of AOX emissions and the reduction of COD in the environment. Therefore, this study is carried out to investigate the effect of the HRT on the performance of a combined biological and adsorption processes using a lab-scale GAC-SBBR to simultaneously remove AOX and COD from real paper mill wastewater.

2. Material and methods

2.1. Analytical chemicals

Standard compounds of PCP, C_6HCl_5O (Molecular weight, $M_w = 266$ g/mol; purity 98.9%), and 2,3,4,5-tetrachlorophenol (2,3,4,5-TeCP), $C_6H_2Cl_4O$ ($M_w = 193$ g/mol), were purchased from SUPELCO (USA), while the other standard compounds, such as 2,4,6-trichlorophenol (2,4,6-TriCP), $C_6H_3Cl_3O$ ($M_w = 197.45$ g/mol; purity 97%), 2,4-dichlorophenol (2,4-DCP), $C_6H_4Cl_2O$ ($M_w = 163$ g/mol; purity 98%), and 2-chlorophenol (2-CP), C_6H_5ClO ($M_w = 128.56$ g/mol; purity 98%) were purchased from MERCK-Schuchardt (Germany). Standard solutions containing 2-CP, 2,4-DCP, 2,4,6-triCP, 2,3,4,5-TeCP and PCP were prepared for the standard curve calibrations.

2.2. Real wastewater samples

Real wastewater was collected from a recycled paper mill in Kajang, Malaysia. The point of wastewater collection was the outlet of the dissolved air floatation (DAF) of the paper mill wastewater treatment facilities at the location. Wastewater was collected once a month and kept in the freezer for further use. The characteristics of the wastewater samples are listed in Table 1. The average COD was 1152 ± 93 mg/L while the average AOX values were 249 ± 15 μg/L (2-CP), 98 ± 8 μg/L (2,4-DiCP) and 42 ± 6 μg/L (2,3,4,5-TeCP).

Table 1
Characteristics of the wastewater samples.

Parameters	
COD (mg/L)	1152 ± 93
pH	7.10 ± 0.5
SS (mg/L)	40–90
2-CP (μg/L)	249 ± 15
2,4-DiCP (μg/L)	98 ± 8
2,4,6-TriCP (μg/L)	0
2,3,4,5-TeCP (μg/L)	42 ± 6
PCP (μg/L)	0
Total Chloride Ion (mg/L)	44.28 ± 2

2.3. Operational conditions of the lab-scale GAC-SBBR

A lab-scale GAC-SBBR with a capacity of 6 L and a working volume of 66.7% (4 L) was used in this on-going study as shown in Fig. 1. The reactor was packed with 80 pieces of cylindrically shaped plastic media with GAC of sizes in the range of 6–8 mm. These packing materials played two roles, which is as the adsorption medium and as a medium for biofilm growth. The biofilm was attached to the coarse surface of the granular activated carbon. The biofilm consisting of a mixed culture (4000–5000 mg/L of mixed liquor suspended solid (MLSS)) was taken from the activated sludge treatment plant of the same recycled paper mill in Kajang, Malaysia. The media had a total surface area of 1945 m^2 with a diameter of 20 mm.

The operational conditions for the lab-scaled GAC-SBBR had undergone Treatments 1–4 as summarized in Table 2. The HRT of the reactors was varied starting from 48 h and then subsequently reduced to 24, 12 and 8 h until a steady-state condition was achieved for each HRT. The reactor was operated under aerobic conditions and maintained in suspension with 1 LPM air. The concentration of nutrient supplied to the reactor was based on the aerobic nutrient-spike with a COD:N:P ratio of 100:5:1 (Metcalf and Eddy Inc., 2004).

2.4. Data collection and analysis

The 2-CP, 2,4-DiCP, 2,4,6-TriCP, 2,3,4,5-TeCP and PCP concentrations were monitored via the absorbance wavelengths of 220, 254, 273, 284 and 310 nm, respectively, using a UV–vis Spectrophotometer (Perkin Elmer, USA). The influent and effluent samples were later analyzed using high-performance liquid chromatography (HPLC) (Agilent Technologies 1200, USA) with a Jones Genesis 4.6 × 250 mm, 5-μ column (Agilent Technologies, USA). The

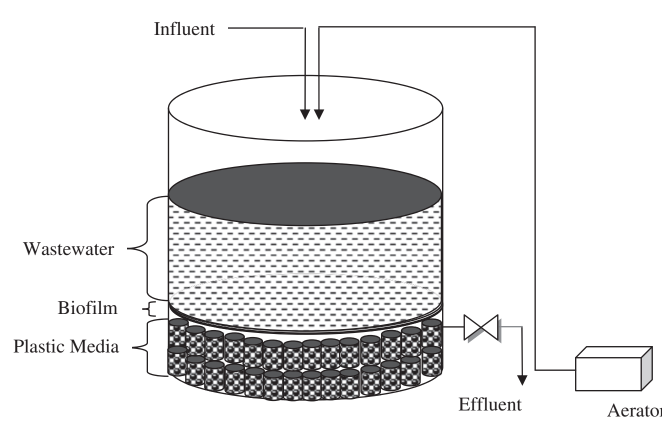

Fig. 1. Schematic diagram of the lab-scale GAC-SBBR.