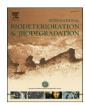


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# Impact of phenol shock loads on the performance of a combined activated sludge-moving bed biofilm reactor system



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#### ABSTRACT

The effect of phenol shock loads (100-3000 mg phenol/L) on the performance of an activated-sludge (AS) - moving bed biofilm reactor (MBBR) system was assessed. The AS-MBBR system could well withstand phenol shocks up to 500 mg phenol/L (organic load of 2.72 kgCOD/(m<sup>3</sup>d)), above which its performance was disturbed, more or less intensely depending on the phenol load. Nevertheless, acclimation of microorganisms to the increasing phenol levels was observed, and full phenolic COD removal was reached at 2000 mg phenol/L. Ammonium removal occurred by both bacterial assimilation and nitrification. Nitrifiers showed higher sensitivity to phenol than heterotrophs, being already impaired at 250 mg phenol/L. However, further adaptation of ammonium oxidizers allowed stable ammonium oxidation activity to be reached up to 1500 mg phenol/L. Nevertheless, nitrite oxidizers were severely affected above 250 mg phenol/L, leading to nitrite build-up. At 3000 mg phenol/L, phenol toxicity strongly limited microbial activity. Meanwhile, COD and ammonium removal performance was dramatically impaired. Moreover, the increasing phenol loads led to the development of filamentous organisms, deteriorating the biomass settling properties. The MBBR, employed downstream of the AS reactor, was frequently subjected to low (sometimes nil) phenol loads, not being therefore acclimated to this compound. Once phenol reached this reactor during some shocks, it accounted for less than 23% of the overall COD removal. The results indicated that the use of a non-acclimated biofilm reactor as a polishing step under phenol stress conditions did not always render improvements to the overall treatment and its implementation is only recommended under certain shock loads.

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#### 1. Introduction

The wastewaters generated by several industrial facilities (e.g., petroleum, chemical, leather processing, textile and pharmaceutical industries) are commonly constituted by phenolic compounds (Tziotzios et al., 2008; Wang et al., 2014). Due to its toxicity, phenol is regarded a priority environmental pollutant in many countries (Huang et al., 2014). In some cases, the concentrations of phenol in the wastewater can reach up to 3000 mg/L (Banerjee and Ghoshal, 2010). At high concentrations, phenol may be toxic to aquatic life in the receiving waters. Besides, it can adversely affect the performance of biological wastewater treatment processes (Xu et al., 2016). If the microorganisms are not properly acclimated to phenol, the biological treatment tends to fail, as reported in

previous studies (Buitrón and Capdeville, 1995; Bajaj et al., 2008; Acikgoz and Ozcan, 2016). Therefore, many efforts have been put towards the development and application of effective treatment strategies to deal with high phenol loads.

Many authors have studied the treatment of phenolic waste-water by specialized strains (Kobayashi et al., 2012; Maza-Márquez et al., 2013), although this approach is not easily applicable to large scale systems (Liu et al., 2016). In general, the conventional activated sludge (AS) process is successfully employed for phenol removal from wastewaters under stable process conditions (Leong et al., 2011). In AS-based processes, heterotrophic and nitrifying bacteria are present in the same reactor tank, so that chemical oxygen demand (COD) and ammonium removal can be simultaneously accomplished. As nitrifying organisms are much more sensitive to inhibition by a wide range of compounds, including phenol (Blum and Speece, 1991; Juliastuti et al., 2003; Kim et al., 2006), any eventual phenol shock load may potentially hamper the nitrification process. Several studies have addressed the

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treatment of phenol-containing wastewaters by the AS process (Yamagishi et al., 2001; Amor et al., 2005; Tziotzios et al., 2008; Papadimitriou et al., 2009; Wang et al., 2014; Tan et al., 2016). Yamagishi et al. (2001) investigated the simultaneous removal of ammonium and phenol in a single-stage AS process. Complete phenol removal was obtained and ammonium was simultaneously nitrified to nitrate. Amor et al. (2005) studied the biodegradation of phenol and its effect on the nitrification process. Phenol was completely biodegraded at concentrations from 100 mg/L to 2500 mg/L in batch assays. However, the increase in initial phenol concentration led to a decrease in the nitrification efficiency. Wang et al. (2014) observed complete removal of phenol (400-1200 mg/ L) in an aerobic sequencing batch reactor (SBR) under high salinity. Evaluating the phenol biodegradation kinetics by olive pulp bacteria in suspended growth reactors, Tziotzios et al. (2008) reported an inhibitory effect at higher phenol concentrations. Papadimitriou et al. (2009) compared the performance of a conventional AS reactor and a SBR for the treatment of wastewaters containing phenol and cyanides, with the latter exhibiting a better performance. Tan et al. (2016) evaluated the treatment of an industrial phenolic wastewater with high salinity using a marine activated sludge containing phenol-tolerant microorganisms. High phenol (99%), COD (80%) and ammonium (68%) removals were obtained by the phenol-acclimated microorganisms.

Other studies have investigated the treatment of phenol-containing wastewaters by biofilm-based technologies, such as the moving-bed biofilm reactor (MBBR) (Hosseini and Borghei, 2005; Li et al., 2011; Hou et al., 2014). Due to its compactness, this process is readily applied to existing plants and has many advantages such as stability to hydraulic and toxic shock loads (Hosseini and Borghei, 2005), high removal efficiency of organic matter and nutrients from a wide range of wastewaters, no need for sludge recirculation, and flexibility of shape and operating load (Ødegaard et al., 2004). Given the challenges involved in the treatment of phenol-containing wastewaters, the MBBR technology is presented as a promising alternative to be combined with an AS reactor to treat waste streams containing phenol.

Overall, satisfactory results in terms of phenol degradation were reported for suspended and attached biomass processes, especially those gradually acclimated to this compound (Fang et al., 2013). The acclimation step is actually reported to be indispensable for the microorganisms to withstand high phenolic loads (Xu et al., 2016). In fact, most of the previous studies investigated the performance of biological treatment systems under gradually increasing phenol loads. However, little attention has been paid on the impact of phenol shock loads (i.e., abrupt changes in phenol concentrations) on both organic matter (COD) removal and nitrification, a situation which is likely to occur in many industrial wastewater treatment plants (Galil et al., 1988). Under these conditions, the operation of the biological treatment facility can be severely disrupted (Galil et al., 1988), and therefore the desired effluent quality may not be reached, even when pre-acclimated microbial consortia are used. Moreover, most studies rely on the application of a single biological reactor, either based on dispersed or attached growth, whereas the combination of both for the treatment of wastewaters with high phenol concentrations has not been reported so far.

Therefore, this research was intended to evaluate the effect of increasing phenol shock loads (100–3000 mg phenol/L) on the performance of a treatment system composed of an AS reactor followed by a MBBR. Special focus was given to the role played by each reactor in the overall treatment process on a long-term basis (around 300 days) and to the impact of the phenol shock on organic matter and nitrogen conversions performed by suspended and attached bacteria. The importance of applying a post-treatment step under phenol stress conditions is also discussed and

recommendations on how to handle shock events are provided.

#### 2. Materials and methods

#### 2.1. Experimental set-up

A combined AS-MBBR system was run in continuous mode for the treatment of a phenolic wastewater. The scheme of the experimental unit is shown in Fig. 1. The AS reactor, with a useful volume of 1.2 L, was composed of two different compartments, an aeration basin and an internal clarifier. This configuration was chosen to better control the sludge retention time (SRT) at around 20 days by manually removing surplus sludge from the aeration compartment (hydraulic SRT control). In such a small-scale device, sludge recirculation was found to be troublesome due to blockage of the silicone tubing due to biomass (sludge) deposition. A porous diffuser was placed at the bottom of the aeration basin for air supply and sludge mixing. The hydraulic retention time (HRT) of the AS tank was set at 12 h. The MBBR, functioning as a post-treatment step for the effluent of the AS reactor, consisted of cylindrical glass vessel with a useful volume of 0.3 L. Kaldnes K5 carriers were used as support material at a media filling fraction of 30%. The HRT was set at 3 h, so that the overall HRT of the AS-MBBR system was 15 h. The reactors were inoculated with activated sludge from a municipal wastewater treatment plant (Alegria/CEDAE, Rio de Janeiro, Brazil).

The combined AS-MBBR system was fed by means of a peristaltic pump with a synthetic wastewater with the following composition: NaCl (1000 mg/L): sodium acetate (463.5 mg/L): methanol (220 mg/L); NH<sub>4</sub>Cl (191.1 mg/L); KH<sub>2</sub>PO<sub>4</sub> (25 mg/L); sodium bicarbonate (350 mg/L); MgSO<sub>4</sub>·7H<sub>2</sub>O (10 mg/L) and trace elements (Vishniac and Santer, 1957). Under normal operating conditions (NC), phenol was added to the influent medium at a concentration of 25 mg/L. Several short-term phenol shock loads were applied to the AS-MBBR system. Throughout the shock periods, influent phenol concentration was abruptly increased from 25 mg/L (NC) to 100 mg/L (1st shock), 250 mg/L (2nd shock), 500 mg/L (3<sup>rd</sup> shock), 1000 mg/L (4<sup>th</sup> shock), 1500 mg/L (5<sup>th</sup> and 6<sup>th</sup> shocks), 2000 mg/L (7th and 8th shocks) and 3000 mg/L (9th and 10<sup>th</sup> shocks). The COD loading rate increased proportionally to the amount of phenol added during the shock tests (Table 1). The last three phenol shock loads were repeated for data validation. Each shock period was maintained for 3 days, after which the system was subjected to normal operating conditions (NC) until a pseudo steady state was re-established (i.e., stable conversions were achieved). This period is referred to as recovery time, whose duration varied depending on the phenol shock load applied. As the first chemical analyses for performance evaluation were carried out around a week after the third day of each shock, the recovery time (in days) could not be precisely determined. Therefore, it was expressed in a week timeframe. The entire operating time of the AS-MBBR system was 293 days.

#### 2.2. Batch experiments

Nitrification activity was assessed by means of batch tests. The experiments were carried out during normal operating conditions (NC) and after each phenol shock. The influent pump was stopped and the reactors were allowed to operate in batch regime. The procedure consisted of adding a pulse of 4 mL in the AS reactor and 1 mL in the MBBR of an ammonium concentrated solution in order to achieve 50 mgNH<sub>4</sub>-N/L at the beginning of the experiment. Subsequently, samples were regularly collected from each reactor at defined time intervals and then filtered through a 0.45  $\mu$ m pore size membrane for determination of ammonium concentrations. The specific ammonium oxidation rate was estimated by linear

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