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## Influence of carrier filling ratio on the performance of moving bed biofilm reactor in treating coking wastewater



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#### HIGHLIGHTS

- The optimal carrier filling ratio of MBBR was investigated.
- The relationship between OURa and NH3-N was simulated by inhibition kinetics model.
- The diversity of microbial communities was affected by carrier filling ratio.

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#### ABSTRACT

This study aims to evaluate the effect of carrier filling ratio on the performance of a moving bed biofilm reactor in degrading chemical oxygen demand, phenol, thiocyanate, and ammonia from coking wastewater at 20 h of hydraulic retention time. The operational experiments under different carrier filling ratios ranging from 20% to 60% were investigated. The maximum removal efficiency of 89%, 99% and 99% for COD, phenol and thiocyanate, and minimum sensitivity to the increasing contaminants concentration in the influent were achieved at 50% carrier filling ratio. The Haldane competitive substrate inhibition kinetics model was used to describe the relationship between the oxygen uptake rate of ammonium oxidizers and the concentration of free ammonium. The highest biofilm microbial community functional diversity (Shannon's diversity index, H') and evenness (Shannon's evenness index, E') were obtained at 50% carrier filling ratio in all runs using a Biolog ECO microplate.

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#### 1. Introduction

As a typical industrial wastewater, coking wastewater contains complicated contaminants and significant amounts of toxic compounds, such as phenolic compounds, thiocyanate (SCN<sup>-</sup>), cyanide (CN<sup>-</sup>), and ammonia (TNH<sub>3</sub>) (Chen et al., 2012; Fang et al., 2013; Oulego et al., 2014; Wu and Zhu, 2012; Yang et al., 2013; Zhang et al., 2012). Activated sludge is a prevalent biological process in treating coking wastewater after the pretreatment which is composed of phenol solvent extraction and ammonia stripping (Guo et al., 2011; Liu et al., 2012; Wang et al., 2012). However,

nitrifying bacteria and specialized microbes are easily washed out because their growth is inhibited by the presence of refractory and toxic compounds (Kim, 2013; Lv et al., 2011; Wang et al., 2013; Zhang et al., 2013a). Poor settleability problems often occur when activated sludge is operated under high loading rates (Jeong and Chung, 2006). Subsequently, the effluent of activated sludge has difficulty in meeting the requirements of the Discharge Standard of Water Pollutants for Iron and Steel Industry (GB 13456-92).

Moving bed biofilm reactors (MBBRs) are advantageous in treating coking wastewater without the problems of activated sludge (Rodríguez-Hernández et al., 2014). MBBRs operate as continued operation biofilm reactors that can accumulate high concentrations of active biomass via microorganism immobilization without backwashing or sludge return. Poor settleability could not damage the operation, and nitrifying bacteria and specialized microbes with low specific growth rates could adhere to the carrier to avoid being easily washed out (Jing et al., 2009; Jin et al., 2013; Zhang et al., 2013b). Jeong and Chung (2006) reported that MBBRs with

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anoxic-oxic-anoxic-oxic reactor arrangement at 3.4 d of hydraulic retention time (HRT) demonstrate good removal performance in treating coking wastewater. Moreover, single-stage MBBRs can be used for the simultaneous removal of chemical oxygen demand (COD), phenols, SCN<sup>-</sup>, and TNH<sub>3</sub> in coking wastewater at 48 h of HRT (Li et al., 2011a). However, little is known about the effect of carrier filling ratio, an important parameter affecting the performance of MBBR in treating coking wastewater. Previous studies adopted high HRTs (Li et al., 2011a), which require large reactor volume and thus high infrastructure investment.

Biofilm formation in MBBRs is a dynamic process. In a steady-state biofilm, the attachment and growth processes reach a balance with the detachment process (Eldyasti et al., 2013; Liu et al., 2003; Walter et al., 2013). The carrier filling ratio could influence both sides of the balance (Wang et al., 2005). On the one hand, more carriers can provide more sites for the attachment and growth of microorganisms. On the other hand, an increase in carrier filling ratio could increase particle–particle collision and promote aeration flux to fluidize carriers, which could enhance the shear stress on the biofilm. Moreover, high aeration flux increases the operational cost (Anderson et al., 2013). Therefore, the carrier filling ratio of MBBR in treating coking wastewater must be optimized to meet the pollution removal requirements and to achieve suitable costs.

This study investigated the effect of carrier filling ratio on the pollutant removal rates at 20 h of HRT was evaluated. The HRT used in this study was shorter than that used previous studies. Moreover, the oxygen uptake rate of the biofilm at different carrier filling ratios was discussed, and the diversity of the microbial communities was analyzed.

#### 2. Methods

#### 2.1. Coking wastewater

The coking wastewater used in this study was collected from the full-scale wastewater treatment facility of a coking plant in Jiyuan, China. The wastewater had been previously subjected to a stripping treatment to facilitate biological treatment. The concentrations of the main pollutants in the wastewater were as follow: 2000–2050 mg/L of COD, 500–540 mg/L of phenol, 540–560 mg/L of SCN<sup>-</sup> and 160–190 mg/L of ammonia.

#### 2.2. Set-up and operation of the MBBR

The MBBR was made of Plexiglas and consisted of an 8 L aeration basin. The aerators were fixed at the center of the bottom of the aeration basin to provide oxygen to the water and to fluidize the biofilm carriers. The airflow rate was measured by a calibrated rotameter and the minimum airflow rate for carrier fluidization was used for each reactor. The dissolved oxygen concentration (DO) was monitored using a polarographic oxygen electrode (HI9142/10, Hanna World Instruments Co., Ltd) and the value of DO was above 3.0 mg/L by using the minimum airflow rate throughout the experiments for each reactor. The carrier was made of polyethylene with a density of about 0.97 g/cm<sup>3</sup> and consisted of a cylinder 10 mm in height and 10 mm in diameter with cross inside. Five sets of MBBRs were concurrently operated. Each reactor had different carrier filling ratios of 20%, 30%, 40%, 50%, and 60%. The HRT of all reactors was sustained at 20 h. During the experiments, the water temperature was controlled at values of  $30 \pm 1$  °C by an immersion thermostat.

All reactors were initially seeded with the same sludge obtained from the aeration tanks of the coking wastewater treatment plant and were ran as continuous flow systems. All reactors were fed with the coking wastewater diluted with tap water to maintain the concentration of COD at approximately 500 mg/L during the start-up period. The influent COD concentration of the MBBR was selected as the operating parameter because of the fluctuation in the composition of the coking wastewater. This parameter was controlled by adding tap water into the coking wastewater. Phosphate was added at a concentration of 0.2 g  $\rm K_2HPO_4/L$  to supply phosphorus for the growth of microorganisms. Bicarbonate was added into the feed wastewater (2 g NaHCO<sub>3</sub>/L) as inorganic carbon source for autotrophic microorganisms to favor nitrification. The pH in the reactor ranged between 7.1 and 8.3 during the experiments.

#### 2.3. Analytical methods

During continuous operation, samples were collected from the influent and effluent of the MBBR daily and were immediately analyzed after filtration through a 0.45 um filter paper. The contents of total suspended solids of suspended growth biomass (TSS<sub>sus</sub>), soluble COD, phenols, and NH<sub>4</sub>-N were measured according to Standard Methods for Water and Wastewater Examination (APHA, 1998). The content of SCN<sup>-</sup> was measured by the ferric colorimetric method. The content of total suspended solids of biofilm biomass (TSS<sub>att</sub>) was measured using the method reported by Li et al. (2011a). Biofilm activity was determined by the oxygen uptake rate (OUR) method (Joanna et al., 1996). For each measurement, 20 carriers and 100 ml supernatant of effluent were used. Activities of the total biofilm and ammonium oxidizers were represented as OUR<sub>t</sub> and OUR<sub>a</sub>, respectively. The specific oxygen uptake rate (SOUR) was expressed based on TSSatt. Throughout out the experiments, OURt was tested when a pseudo steady state was achieved in each stage, while OURa was tested more times when phenol and SCN- concentrations decreased to normal level.

#### 2.4. Biolog methods

Biolog ECO microplates (Biolog, Inc.), which are plastic microtiter plates with 96 wells, were used in this study. Each plate consisted of three replicates (comprising 31 sole carbon sources and one water blank). In this study, 5 g (wet weight) of carriers from each MBBR was suspended in 45 mL of buffered saline solution (0.9% NaCl, pH 7) and shaken four times (15 s each) using an ultrasonic cleaning machine (Baoding Leici Co., 40 kHz, 250 w). A 1 mL aliquot of the resulting suspension was diluted with additional buffered saline solution to control the optical density (OD) close to 0.05 at 600 nm and ensure that the five sample solutions contained approximately the same biomass concentration. Then, the suspension was used to inoculate the Biolog plates (150 µL per well). The plates were incubated at 30 °C, and absorbance at 595 nm was measured using a microplate reader (BIO-RAD Model 550) every 12 h up to 180 h. The reading of each well of the Biolog plates was corrected by subtracting the value of the water blank. The overall color development, expressed as average well color development (AWCD), was calculated as the mean of the blanked absorbance values for all 31 wells per reading time. AWCD =  $\sum$ (C-R)/N, where C is color production in each well (OD measurement), R is the absorbance value of the control well, and N is the number of substrates (ECO plates, N = 31). Three replicates per treatment per sampling site were performed. The functional diversity, as measured by Shannon's diversity index (H'), was calculated using the equation  $H' = -\sum P_i (\log P_i)$ , where  $P_i$  is the ratio of the blanked absorbance value of each well to the sum of absorbance values of all wells. The functional evenness, as measured by Shannon's evenness index (E), was calculated using the equation E = H'log S, where S is the number of available carbon sources, the absorbance of which is more than 0.2 (Calderón et al., 2012; Huang et al., 2008; Kong et al., 2013).

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