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Performance evaluation of attached biofilm reactors for the treatment of wastewater contaminated with aromatic hydrocarbons and phenolic compounds

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ABSTRACT

The present study investigated the performance of single stage moving bed biological reactor (MBBR) and submerged aerated biological filter (SABF) for the treatment of wastewater contaminated with heterocyclic and homocyclic aromatic hydrocarbons along with phenolic compounds commonly discharged from coal and biomass gasification plants. Performance evaluation of both the bioreactors was carried out by varying hydraulic and organic loading rates (OLR). Removal efficiencies (R.E) of 90.4 ± 0.78% and 85.8 ± 1.96% were achieved in MBBR and SABF, respectively at HRT of 24 h and OLR of 2.45 kg/m^3/day. Increasing the OLR to 4.77 kg/m^3/day resulted in the reduction of R.E of MBBR and SABF to 86 ± 0.96% and 77.8 ± 1.45%, respectively. MBBR showed better stability against hydraulic and organic shock loads in comparison to SABF. The effect of co-contaminants such as phenol and cresol on overall reactor performance was also investigated. The coexistence of phenol (300 mg/L) and cresol (100 mg/L) affected the removal of other hydrocarbons and resulted in accumulation of the metabolic intermediates. TOC R.E was 86.8% and 84.9% while reduction in toxicity was 61% and 57% in MBBR and SABF during simultaneous treatment of mixed hydrocarbons and phenolic compounds (OLR = 3.43 kg/m^3/day, HRT = 24 h). Modified Stover- Kincannon model was incorporated to elucidate the substrate utilization kinetics. Characterization of attached biofilm on the carrier elements showed a significant variation in extracellular polymeric constituents with different pollutant concentrations.

1. Introduction

Complex industrial wastewater generated from different industrial activities such as coal gasification, coke oven, petroleum refineries and biomass gasification contains mixture of toxic organic and inorganic pollutants which are often, either partially treated or untreated before being discharged into the environment. The primary constituents present in these wastewater streams include several toxic contaminants such as nitrogen, sulphur and oxygen containing heterocyclic hydrocarbons, polycyclic and monocyclic aromatic hydrocarbons, different phenolic compounds and ammonia [1,2]. COD of coal gasification and coking wastewater was reported to be as high as 12,500 mg/L [3]. Shi et al. [1] and Zhu et al. [2] reported that major constituents present in raw coking wastewater includes phenols (227 ± 70.3 mg/L), pyridine (43.2 ± 8.6 mg/L), quinoline (23.9 ± 9.6 mg/L), naphthalene (1.6 ± 0.8 mg/L), carbazole (10.9 ± 3.1 mg/L), dibenzofuran (11.7 mg/L) and dibenzothiophene (10.8 mg/L) contributing to the total COD of 1766–1937 mg/L. Most of these pollutants are carcinogenic and mutagenic in nature and this hazardous effluent induce several adverse effects in the receiving environmental bodies, if not treated properly.

Application of different biological processes for the treatment of coal gasification wastewater including conventional activated sludge process [4], membrane bioreactor [2], upflow anaerobic sludge blanket (UASB) reactor [5] and expanded granular sludge blanket reactor (EGSB) [6] have been extensively explored in the recent years. Several previous studies [7] report-limitations such as loss of nitrifying bacteria and specialized microbes due to their growth being inhibited by the presence of refractory and toxic compounds, especially in conventional activated sludge processes. Moreover, the reactor performance under high loading conditions lead to poor sludge settleability [8] and UASB and EGSB require longer start-up period (3–8 months). To circumvent the disadvantages of the conventional treatment processes, different advanced biofilm reactors have been developed to achieve simultaneous and reliable removal of organic carbon and nitrogen.

Moving bed biological reactor (MBBR) and submerged aerated biological filter (SABF) are the most commonly used dynamic and static fixed film bioreactor systems, respectively. Presence of bio carriers

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provides high concentration of micro-organisms that result in high degradation rate, ease of operation, stability under large load variations with relatively low footprint and minimal sludge generation [9]. Application of MBBR and SABF has been extensively investigated for the removal of various toxic pollutants such as pharmaceutical compounds [10], 2,4,6 trinitrotoluene [11], emerging contaminants like benzotriazole and benzothiazoles [12], diethylphthalates and diallylphthalates [13]. Despite biofilm reactors being the key technology for the removal of toxic and emerging pollutants, only very few studies have explored the use of single stage MBBR and SABF for simultaneous removal of toxic contaminants present in coal gasification and petroleum refinery wastewater [14–16]. Studies by Li et al. [15] reported the ability of MBBR to resist shock loading on increasing phenol concentration. Shi et al. [16] reported an efficiency greater than 97% for mixed aromatic hydrocarbons and phenols in bio-augmented biological aerated filter. Most of the existing studies reported the reactor performance either in terms of removal of overall COD or phenolic compounds. These studies did not provide a clear understanding about the degradation and toxicity of individual constituents of the wastewater. Studies pertaining towards the simultaneous removal of mixed aromatic hydrocarbons with phenolic compounds are very limited. Moreover, these studies reported the use of either of these bioreactors for the treatment of pollutants individually or in mixtures. Owing to the differences in the reactor configuration and the carrier media movement, both the reactors may respond differently to varying operation conditions. To the best of our knowledge there is no previous published literature, detailing and comparing the performance of these two bioreactors for the simultaneous removal of mixed pollutants and reduction in toxicity. The information on the reactor stability and resilience under various organic and hydraulic loading rates and shock loads are crucial parameters to evaluate the system efficacy. However, not many studies are being carried out on these aspects with respect to MBBR and SABF treating complex mixed pollutants. Furthermore, limited studies have investigated the effect of these toxic compounds on the biofilm characteristics in terms of extracellular polymeric substances (EPS) and their variation in concentration and composition during the treatment. The present study thus provides valuable information to comprehend the biofilm formation, adhesion and regulation during the treatment of such complex wastewater streams.

The objectives of the present study are to: (i) evaluate and compare the performance of MBBR and SABF for the treatment of wastewater contaminated with several aromatic hydrocarbons and phenolic compounds under different operating conditions; (ii) study the effect of different operational parameters such as HRT, OLR, and toxic shock loads on the performance; (iii) assess the reactors performance in terms of toxicity reduction of treated effluent; (iv) investigate the variation in attached biofilm composition in terms of extracellular polymeric substances (EPS) during different operational stages and (v) determine the substrate removal kinetics.

2. Materials and methods

2.1. Microbial inoculum and culture medium

The MBBR and SABF reactor was inoculated with mixed microbial culture which is previously acclimatized with naphthalene [17]. Some of the prominent bacterial strains identified in mixed microbial consortium were *Chryseobacterium* sp. and *Rhodobacter* sp. [17]. Following composition of minimal salt medium (MSM) was used for the preparation of synthetic wastewater (g/L): $Na_2HPO_4{\cdot}2H_2O$ (3.0), KH_2PO_4 (1.5), $(NH_4)_2SO_4$ (0.5), $MgCl_2{\cdot}6H_2O$ (0.1), $CaCl_2$ (0.02), and trace element solution 1 mL/L [18]. The pH of the MSM was 7.0- 7.2. Synthetic wastewater was prepared by using MSM along with different pollutants which include heterocyclic pyridine, quinoline, benzothiophene, benzofuran; benzene (monocyclic); naphthalene (polycyclic) aromatic hydrocarbons and phenolic compounds (Phenol and Cresol) at varying initial concentrations. Chemical structures of different target contaminants are given in ESM (Table S1).

2.2. Experimental studies

2.2.1. Experimental setup and operation: continuous biofilm reactors

(a) Submerged aerated biological filter (SABF)

A plexi glass reactor with a cylindrical shape was used as SABF reactor. The outer diameter and height was 10 cm and 50 cm respectively. SABF reactor was packed with the open pore spirals (Fujino spirals, India) having a surface area of approx 350 m^2/m^3. Effective media height inside the reactor was 30 cm and all the carrier elements were completely immersed in MSM. Initial porosity of the packed bed was 0.88. Sampling ports were also provided along the length of the media bed with an equidistant spacing of 14 cm. The outlet port for the collection of effluent was provided 5 cm above the top layer of the carrier media. Air diffuser was provided at the bottom of the reactor and air (1 L/min) was supplied using an air pump (Buoy, China). Flow rate was measured using an air flow meter (Placka Instrumentation, India). The circular base plate with perforations was provided at the bottom of the reactor to support the carrier media. U-tube manometer, filled with water, was used to measure the pressure drop across the bed height of the reactor. Schematic of the SABF reactor is shown in Fig. 1(a).

- Moving Bed Biological Reactor (MBBR)

The MBBR reactor was fabricated using cylindrical plexiglass column with an outer diameter of 10 cm and total height of 50 cm. The effective volume of the reactor was 2.3 L. The carrier media used in MBBR reactor was made of polyethylene with an internal diameter of 20 mm and density of about 0.92 g/cm^3. The suspended carrier media had a total projected surface area of 402 m^2/m^3 and was cylindrically shaped with the fins outside and projections inside. The suspended carrier filling ratio was about 16% to ensure homogenous circulation of media inside the reactor with air passage introduced at the bottom. Coarse bubble aeration was provided through diffuser from the bottom of the reactor using an air pump at 1 L/min flow rate, during the start-up phase. The circular base plate with perforations provided above the air diffuser was designed in such a way that air could be supplied in the reactor to ensure complete hydraulic circulation of the carrier media. The schematic of MBBR is shown in Fig. 1(b).

The efficiency of both the reactors was evaluated under different operating conditions. The synthetic wastewater containing various target pollutants was pumped to the reactor with a peristaltic pump (PP30, Miclins, India) through the inlet port placed at a distance of 5 cm from the bottom of the reactor.

A glass container of 5L volume fitted with air tight Teflon septum was used as an influent tank with a magnetic stirrer to ensure homogenous mixing. The influent pH was between 7.0–7.2. The liquid samples were collected everyday from both the inlet and outlet ports for residual pollutant concentration analysis. TOC, residual toxicity, pH, DO and suspended solids concentration of the treated wastewater were periodically measured. Biomass concentration, porosity and EPS constituents of the biofilm were determined periodically during different operational stages.

2.2.2. Start up and performance evaluation of MBBR system

During the initial start-up, both the reactors were seeded with bacterial inoculum to favor the early attachment of micro-organisms and thus facilitate biofilm formation on the carrier media. For this purpose, the enriched microbial inoculum was circulated in the reactor at a flow rate of 0.2 L/h and the reactor was operated in closed loop condition. This was continued for an initial period of 20 days to allow