Biochemical Engineering Journal 43 (2009) 98–105

Contents lists available at ScienceDirect

Biochemical Engineering Journal

journal homepage: www.elsevier.com/locate/bej

Biotrickling air filtration of 2-chlorophenol at high loading rates

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ARTICLE INFO

Article history:
Received 9 February 2007
Received in revised form 4 September 2008
Accepted 5 September 2008

Keywords:
Air pollution control
Trickling biofilter
Pseudomonas pickettii
Ceramic packing
Inhibition
Kinetics

ABSTRACT

The degradation of 2-chlorophenol vapours in air was performed in a trickling biofilter packed with ceramic material seeded with the bacterium *Pseudomonas pickettii*, strain LD1. The system performance was evaluated under varying operating conditions (inlet 2-chlorophenol air concentrations from 0.10 to 3.50 g m^{-3}, and superficial air velocities of 30.0, 60.0, and 120.0 m h^{-1}). For all air velocity the maximum degradation rate was obtained for loading rates of 40 g m^{-2} h^{-1}. Higher loading conditions resulted in strong inhibition of microbial activity, particularly severe at high air velocity. Process analysis, performed using data on pollutant concentration profiles along the filter packing obtained under different conditions of inlet concentration and air velocity, proves that best performance (i.e. maximum degradation efficiency and capacity) can be obtained for a narrow range of operating conditions, which can be ensured by proper design of biofilter size (i.e. diameter and height). Kinetic analysis of experimental data confirms that 2-CP inhibits microbial activity in the biofilter bed. Experimental data are satisfactorily fitted by the Haldane kinetic equation up to a critical value of loading rate, beyond which the experimental degradation rate is overestimated by the kinetic model. The inhibition appears to be affected by the loading rate, and the estimated inhibition constant linearly increases with increasing empty bed residence time.

1. Introduction

Chlorinated phenols are xenobiotic compounds used in industrial processes to manufacture pesticides, herbicides, fungicides, disinfectants, antiseptics, wood and glue preservatives, paints, solvents, and pharmaceuticals.

2-Chlorophenol (2-CP) along with, 2,4-dichlorophenol, 2,4,6-trichlorophenol and pentachlorophenol are listed by the US Environmental Protection Agency as priority environmental pollutants [1], toxic and carcinogenic to man [2,3].

Chlorophenols enter the environment as industrial effluents, most notably in gaseous and liquid waste streams from herbicide and biocide production plants [4]. Treatment of water and air streams polluted by chlorophenols is carried out by chemical methods, including supercritical [5], wet air [6], photocatalytic [7], and Fenton oxidation [8], or catalytic hydrodechlorination in gas or liquid phase [9,10]. These processes require either severe operating conditions [5,6] and dosage of chemicals [8–10] or complex reactor configurations [7], which make biological methods an attractive alternative for the removal of chlorinated phenols from waste streams.

Biological oxidation of chlorophenols is performed by specific bacterial strains belonging to the genera *Pseudomonas* [11], *Alcaligenes* [12], and *Rhizobium* [13], and fungal strains belonging to the genera *Phanerochaete* [14]. Aerobic degradation takes place via formation of catechols, while anaerobic degradation occurs via reductive dechlorination [15]. *Pseudomonas pickettii*, strain LD1, is reported to mineralize 2-CP, 3-CP and 4-CP through the chlorocatechol pathway [16]. Another strain obtained from the same mixed culture from which LD1 was isolated, i.e., *Pseudomonas putida*, strain CP1, displays similar biodegradation pathways [17]. However, LD1 strain shows higher tolerance to high and variable 2-CP concentration than CP1 strain [17–19]. Degradation of 4-CP was demonstrated in a fluidized bed bioreactor loaded with yeast (*Candida tropicalis*) immobilized onto granular activated carbon [20] and in an up-flow sludge blanket reactor under methanogenic conditions [21]. Enhanced removal efficiency can be obtained by operating anaerobic and aerobic treatment processes in combination [22].

High concentrations of chlorophenols are usually inhibitory for microorganisms [23–26]. They are reported to increase the duration of the lag phase [27] and decrease the rate of biodegradation of different species of bacteria and fungi [28,29]. The recalcitrance of chlorophenols increases with increasing number of chlorinated susbstituents on phenolic ring [15]. Position of the phenolic ring may also affect the biodegradability of such substances [16].

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doi:10.1016/j.bej.2008.09.007

Nomenclature

a	specific surface area (m^{-1})
C	2-CP concentration in the airflow ($g\,m^{-3}$)
d	diameter (m)
D	dispersion coefficient ($m^2\,s^{-1}$)
H	overall filter packing height (m)
k	specific degradation rate ($g_{2\text{-CP}}\,g_{biomass}^{-1}\,h^{-1}$)
k_G	gas-film volumetric mass transfer coefficient (h^{-1})
K_I	inhibition constant ($g\,m^{-3}$)
K_S	saturation constant ($g\,m^{-3}$)
r	degradation rate ($g\,m^{-3}\,h^{-1}$)
u	superficial velocity ($m\,h^{-1}$)
X	biomass concentration ($g\,m^{-3}$)
z	axial coordinate (m)

Greek letters

Γ	degradation capacity ($g\,m^{-3}\,h^{-1}$)
Λ	loading rate ($g\,m^{-2}\,h^{-1}$)
ε	bed porosity
η	degradation efficiency (%)
τ	empty bed residence time, EBRT (s)

Subscripts

b	biofilm
g	gas
in	inlet
max	maximum
out	outlet
p	particle

Acclimatization of biomass to chlorophenols markedly enhances their ability to degrade such compounds both by reducing the initial lag-phase [27] as well as by countering biomass inhibition [25]. The Haldane equation [30] is one of the most commonly used models for describing the growth kinetics of microorganisms inhibited by chlorophenols, considered either as growth substrates [24] or as nongrowth substrates [26].

The feasibility of air biofiltration was demonstrated at laboratory scale for different inhibitory compounds, including chlorophenols [31] and other volatile organic carbons [32–38]. The gas flow rate and the inlet pollutant concentration are the most important parameters affecting the process performance. For a specific filter bed volume, the gas flow rate is the key parameter for varying the gas residence time, which should be long enough to permit the pollutant and oxygen transfer from the gas phase to the biofilm and their biodegradation by the immobilized microflora. Biofiltration proves to be highly efficient for dilute air streams of easily biodegradable compounds, and less proficient in the treatment of more concentrated emissions of moderately or poorly biodegradable pollutants [39], as high concentrations of recalcitrant compounds may produce inhibition effects on the metabolic activity of the microbial population [40]. Because of inhibition effects on microbial growth rate, the degradation rate of inhibitory compounds in biofilters decreases at high loading rates [31,41], while in the absence of inhibition the biofilter degradation capacity increases at low loading rate, when the process is kinetically limited by the low availability of organic substrate for reaction, and it attains a maximum value determined by the viable reactor biomass concentration [42–46].

In this work, process performance are assessed at high loading rates, with the main objective of evaluating the inhibiting effect of high 2-CP concentrations on degradation capacity in trickling biofiltration of xenobiotic compounds.

2. Materials and methods

2.1. Equipment

The experimental set-up, schematically represented in Fig. 1, comprises a glass column with square cross-section filled with a clay adsorbent (Buzzi Unicem, Italy). The reactor design characteristics are listed in Table 1, while Table 2 reports the properties of the packing material.

The filter packing was supported by a stainless steel punched plate covered by a gravel layer to get a homogeneous distribution of the airflow through the filter packing and proper liquid drainage as well as to prevent clogging of the holes. The bioreactor was provided with four ports for sampling of gas and packing material. All tubes and fittings of the apparatus were made of Teflon in order to minimize adsorption of 2-CP from airflow.

The biofilter was operated counter-currently. The feed airflow laden with 2-CP was generated by using the detailed procedure presented elsewhere [45]. The aqueous phase was continuously recirculated to the top of the bioreactor by a peristaltic pump, and sprinkled on the surface of the filter packing through a perforated diffuser to ensure uniform distribution. The trickled aqueous phase, continuously drained from the column, was collected in a 2-l well-mixed vessel, where the pH was controlled at 8.0 ± 0.2 by addition of a 1N NaOH aqueous solution. Fresh mineral medium was continuously supplied to the biofilter, at a flow rate which was varied from 0.15 up to 4.0 l h^{-1} depending on the organic loading applied.

2.2. Bacterial strain

A pure culture of *Pseudomonas pickettii*, strain LD1, was used to inoculate the trickling bioreactor. LD1 strain is a gram-negative, aerobic, non-sporulating, motile rod-shaped bacterium with polar flagella, capable of completely mineralizing 2-CP by utilizing it as sole source of carbon and energy in a mineral salts medium without supplemental additions of micronutrients or growth cofactors [13,14]. The basal medium used for LD1 growth and in the experimental runs was composed of (in grams per liter of deionized water): $(NH_4)_2SO_4$, 3.0; Na_2HPO_4, 2.5; KH_2PO_4, 1.0; $Ca(NO_3)_2{\cdot}4H_2O$, 0.04; $MgSO_4{\cdot}7H_2O$, 0.2; Fe (III) ammonium citrate, 0.01. LD1 cells were aerobically cultivated as detailed elsewhere [31].

2.3. Experimental procedure

Experiments were carried out at laboratory room temperature (in the range 19–26 °C) under non-sterile conditions. The superficial liquid velocity (6.0 m h^{-1}) was higher than the minimum-wetting rate for the packing material and constant throughout the experimental tests.

To start up the biofiltration process, the trickling biofilter was inoculated using 4.0 l of bacterial suspension (average content of 0.30 g cell dry weight l^{-1}) pregrown on 2-CP. The suspension was poured on top of the reactor packed with sterilized BCA, and recycled through it for 3 days. Continuous operation of the biofilter was then started up at a superficial liquid-flow velocity of 6.0 m h^{-1}, a superficial airflow velocity (V_a) of 15.0 m h^{-1} (corresponding to an EBRT of 4 min), and an influent 2-CP air-phase concentration (C_{in}) of 0.1 g m^{-3}. Fresh mineral salts medium was continuously added to the biofilter at a flow rate of 0.075 l h^{-1}. The bioreactor was run under these conditions to enable gradual microbial attachment and development of a biofilm of active cells around the granules surface.