Bioresource Technology 101 (2010) 8067–8073

Contents lists available at ScienceDirect

Bioresource Technology

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

Treatment of a BTo-X-contaminated gas stream with a biotrickling filter inoculated with microbes bound to a wheat bran/red wood powder/diatomaceous earth carrier

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ARTICLE INFO

Article history:
Received 24 January 2010
Received in revised form 7 May 2010
Accepted 29 May 2010
Available online 23 June 2010

Keywords:
Solid carrier
Microbial consortia
Inocula
BTo-X
Biotrickling filter

ABSTRACT

Microbes bound to a wheat bran/red wood powder/diatomaceous earth carrier were used as inoculants for a biotrickling filter (BTF) for treating gases contaminated with a mixture of benzene, toluene, and *o*-xylene (BTo-X). An overall removal efficiency of more than 87.9% was achieved after a start-up period of as low as 4 days. At BTo-X loading rates (LRs) below 60.0 g/m^3 h, the BTF's performance was similar for EBRTs of 90, 60, 45 and 30 s with an elimination capacity (EC) almost approaching the LR; stable REs above 91.3% for benzene and toluene and above 82.8% for *o*-xylene were achieved. A maximum EC of 97.7 g/m^3 h was obtained at inlet load of 146.4 g/m^3 h. The mass ratio of carbon dioxide produced to the BTo-X removed was approximately 2.62, which confirmed complete degradation of BTo-X. The results demonstrate that microbes bound to a solid carrier can be an alternative to traditional liquid inoculums applied in BTFs and highlight their potential applicability to BTF technologies.

1. Introduction

Biotreatment of VOCs provides an environmentally-friendly and low-cost alternative to other physical and chemical treatment technologies such as incineration, catalytic oxidation, and adsorption (Cox and Deshusses, 2002; Hassan and Sorial, 2009; Lee et al., 2009). The most widely utilized bioreactors for the control of VOCs emissions are biofilters (BFs) and biotrickling filter (BTFs). BTFs work in a manner similar to BFs, except that an aqueous mineral nutrient solution is trickled over the packed bed and that the packing is made of inorganic materials. BTFs facilitate more consistent operation than traditional BFs due to better control of overall pressure drop, nutrient concentration, and pH, and enable higher pollutant elimination rates to be obtained for a broader range of pollutants (Jorio et al., 2000; Arriaga et al., 2006; Sercu et al., 2007; Popat and Deshusses, 2008; Nicolella et al., 2009). Efficient removal of, for example, aromatics, some chlorinated hydrocarbons, organic sulfur compounds, and other VOCs had been reported in lab-scale BTFs. Most reported studies were conducted on single VOCs, while studies on VOCs mixture for biofiltration are relatively scarce (Jorio et al., 2000; Strauss et al., 2004; Cai et al., 2007; Mohammad et al., 2007; Sempere et al., 2008).

In BTFs, few microorganisms are initially present on the inert packing material and inoculation is therefore always needed. The choice of inocula could potentially influence the start-up of a BTF, the removal of pollutants and the reactor stability (Thompson et al., 2005; Sercu et al., 2007; Kumar et al., 2009). In general, BTFs are mainly inoculated with (enriched) mixed microbial consortia (Mohammad et al., 2007; Ryu et al., 2010) or pure cultures (Arriaga et al., 2006; García-Peña et al. 2008; Lee et al., 2009). For industrial application, a mixed culture would be preferred since there is less of a need to prevent contamination with unwanted microorganisms, and the microbial diversity allows for greater flexibility with respect to substrates (Kleerebezem and van Loosdrecht, 2007; Kumar et al., 2009).

Since it can take up to several weeks after start-up before bioreactors effectively remove gaseous pollutants (Sercu et al., 2005), mixing of BTF sludge inoculums with specialized strains for removal of dimethyl sulfide (DMS) was carried out in an attempt to shorten the start-up period (Sercu et al., 2007). The addition of the DMS-degrading strains was beneficial initially, but this effect disappeared largely over the long run. The selection of the inoculums becomes increasingly important when the pollutant is more difficult to degrade (Cox and Deshusses, 2001). Often several weeks to several months are required to obtain adapted inoculums with high removal capacity. If adapted inoculums were available at any time, the BTF technology would be more competitive compared to other treatment methods for VOCs; however, enriched/prepared liquid microbial inoculums may be difficult to preserve over extended time periods and to transportation (Ye et al., 2009).

Therefore, in this study, a microbial consortium bound to a solid carrier was employed as an alternative to traditional liquid inoculums for treatment of VOC gases by BTF. The long-term stability of

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doi:10.1016/j.biortech.2010.05.096

the inoculums was tested, and the start-up times, effect of BT*o*-X loading on the EC of BTF, CO_2 production, and kinetic behavior of the system were determined.

2. Methods

2.1. Preparation of inoculums

Wheat bran, red wood powder, and diatomaceous earth (85%:10%:5%), which were obtained from wheat processing factory of Hangzhou, China, Jinwang timber processing factory of Hangzhou, China, and World Minerals Inc., Celite, China, respectively, were used for the solid carrier for microbial growth. The liquid medium included the following components (per L): K_2HPO_4, 0.11 g; KH_2PO_4, 0.04 g; NH_4Cl, 0.54 g; $MgSO_4$, 0.067 g; $CaCl_2$, 0.036 g; $FeCl_3$, 0.25 mg; $MnSO_4$, 0.03 mg; $ZnSO_4$, 0.04 mg; $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, 0.03 mg.

Fifty g of the solid carrier, 50 mL of the liquid medium and 10 mL of enriched microbial consortium grown on BT*o*-X compounds as the sole carbon and energy source (Ye et al., 2009) were sufficiently mixed in 500 mL air-sealed bottle sealed with Teflon-coated rubber stoppers and aluminum crimps. A sufficient amount of headspace was provided to avoid oxygen limiting conditions. The compounds were added in amounts to obtain an approximate total of 250 mg BT*o*-X per bottle. The incubation was carried out at 35 °C for 48 h. The cultures were dried at 35 °C, and ground into small particles with an agate mortar before storage at 4 °C.

2.2. Biological community structure and activities

The diversity of bacteria in the enriched microbial consortium before and after adsorption onto the solid carrier was analyzed by denaturing gradient gel electrophoresis (DGGE). DNA of the prepared fresh cultures and the enriched microbial consortium was extracted using a Wizard genomic DNA purification kit. Polymerase chain reaction (PCR) to amplify 16S rRNA gene sequences was done as previously described (Zhang et al., 2008). DGGE was performed using a D-Code 16 × 16 cm acrylamide gel system (BioRad, Hercules, CA) following published procedures (Muyzer et al., 1993).

Measurement of biological activities by the carrier-bound microbes after different storage periods was done by oxygen uptake (OUR) determinations in a 2.5 mL vessel fitted with a Clark-type oxygen electrode system (Oxytherm, Hansatech, United Kingdom). Samples were saturated with air and the endogenous respiration rate was monitored for 2–5 min. The BT*o*-X-induced OUR was determined after addition of a BT*o*-X solution to a final concentration of 0.5 mM, and the OUR was corrected for the endogenous respiration. OUR experiments were done in duplicate, and rates were correlated to the dry weight of each sample.

2.3. BTF setup and operational conditions

The BTF was constructed of three cylindrical glass sections with an internal diameter of 12 cm and a total length of 110 cm. The outlet for treated air and the inlet for nutrient feed were located in the head space section and the inlet for BT*o*-X-containing air and filtered nutrient solution were located at the bottom section. The BTF was equipped with three sampling ports to measure BT*o*-X and CO_2 concentrations, located at 20 (inlet port), 60, and 100 (outlet port) cm of column height. Additional ports located at 36, 47, 80, and 92 cm were used for temperature measurement and to recover bed particles for microbial analyses. The BTF was packed with pelletized polyurethane foam (PUF) with a diameter of 14–18 mm to a depth of about 60 cm. The palletized PUF had an initial porosity of 91.0%, and a specific surface area of 380 m^2/m^3. The porosity was measured by the mercury porosimeter (Micromeritics autopore IV), and the specific surface area was determined by the nitrogen adsorption–desorption method.

Gas-phase BT*o*-X were generated individually by passing a known flow of air into troughs containing liquid-phase B, T and *o*-X, and mixing the generating BT*o*-X vapor with another stream of air in a mixing chamber. The resulting BT*o*-X waste gases were introduced through the bottom of the column; flow rate was adjusted using mass flow controllers. The BTF was started by adding 200 g of the prepared inoculums into 2000 mL of basal mineral medium (BMM) composed of (per L): K_2HPO_4, 0.11 g; KH_2PO_4, 0.04 g; NH_4Cl, 0.54 g; $MgSO_4$, 0.067 g; $CaCl_2$, 0.036 g; $FeCl_3$, 0.25 mg; $MnSO_4$, 0.03 mg; $ZnSO_4$, 0.04 mg; $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, 0.03 mg. The BMM solution was continuously recirculated over the packed bed in a counter-current flow with respect to the air flow using a peristaltic pump at a constant volumetric flow rate of 10 L/h. Two-hundred mL of recycled liquid was replaced with fresh mineral medium once a day (average liquid residence time of 10 d). The experiments were conducted at room temperature (20–25 °C). The conditions employed during the experimental runs are summarized in Table 1.

2.4. Analytical methods

BT*o*-X concentrations in the gas-phase were measured in a gas chromatograph (GC, Agilent 6890) fitted with a HP Innowax capillary column (30 m × 0.32 mm × 0.5 μm) and flame ionization detector (FID). Nitrogen was the carried gas at a flow rate of 33.4 ml/min. The oven and detector temperatures were set at 200 and 90 °C, respectively. A GC (Agilent 6890) equipped with a thermal conductivity detector (TCD) was used for determining the CO_2 concentrations in the BTF's inlet and outlet.

Scanning electron microscopy (SEM, Philips, XL30) was used to visualize the morphology of microbial populations on the carrier and the BTF. The biofilm samples were gently washed with 0.1 M phosphate buffer, pH 7, and fixed with 2.5% glutaraldehyde and 1% osmic acid for 4 and 3 h, respectively. The fixed samples were dehydrated by successive passages of 30%, 50%, 70%, 85%, and 95% ethanol solutions and absolute ethyl alcohol. The dehydrated samples were dried with a CO_2 Critical Point Dryer, and were finally placed on metallic stub and covered with a gold layer in a sputter coater to increase their electrical conductivity.

Table 1
Experimental operating conditions of each phase in the BTF for the treatment of a BT*o*-X-contaminated gas stream.

	I	II	III	IV	V
Operational periods (days)	1–16	17–59	60–105	106–149	150–186
B inlet concentration (g/m^3)	0.07–0.13	0.11–0.54	0.10–0.54	0.10–0.55	0.10–0.46
T inlet concentration (g/m^3)	0.08–0.15	0.11–0.51	0.10–0.51	0.11–0.51	0.11–0.41
o-X inlet concentration (g/m^3)	0.05–0.11	0.09–0.51	0.09–0.51	0.09–0.51	0.09–0.42
Total loading rate (g/m^3 h)	8.30–15.60	11.90–62.50	17.60–93.80	24.10–126.00	35.70–154.80
Gas flow rate (m^3/h)	0.389	0.389	0.583	0.778	1.167
EBRT (s)	90	90	60	45	30