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Low-temperature anaerobic biological treatment of toluene-containing wastewater

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ABSTRACT

Two expanded granular sludge bed-anaerobic filter (EGSB-AF) bioreactors, R1 and R2, were operated at 15 °C for the treatment of toluene-contaminated volatile fatty acid-based wastewater. The seed inoculum and the R1 reactor were unexposed to toluene, prior to and during the trial, respectively. Both reactors were operated at a hydraulic retention time of 24 h at applied organic loading rates of 0.71–1.43 kg chemical oxygen demand (COD) $m^{-3} d^{-1}$. Toluene was supplemented to the R2 influent at concentrations of 5–104 mg toluene l^{-1} (solubilised in ethanol). Bioreactor performance was evaluated by COD and toluene removal efficiency, and the methane content of biogas (%). Specific methanogenic activity and toxicity assays were employed to investigate the activity and toluene toxicity thresholds of key trophic groups, respectively, within the seed and reactor biomass samples. COD and toluene removal efficiencies of 70–90% and 55–99%, respectively, were achieved during the 630-d trial. Metabolic assays suggested that a psychrotolerant H_2/CO_2-utilizing methanogenic community developed in the toluene-degrading biomass. The results indicate the viability of low-temperature anaerobic digestion for the treatment of wastewater containing toluene.

1. Introduction

Toluene (C_7H_8) is an aromatic hydrocarbon belonging to the BTEX group of hazardous volatile organic compounds (VOC), which includes benzene, ethylbenzene and xylene. It occurs naturally in the anoxic sediments of lakes (Jüttner and Henatsch, 1986; Fischer-Romero et al., 1996) through the degradation of phenylalanine by several species of anaerobic bacteria (Heider et al., 1999). The use of this, relatively water-soluble, aromatic hydrocarbon as a solvent in the production of paints, thinners, adhesives, inks and many pharmaceutical products, results in its subsistence in air and wastewater emissions associated with these industries. Toluene concentrations in industrial wastewaters vary between 7–753 mg l^{-1} (De Witt, 1999) depending on the manufacture type. In addition, toluene is also found in groundwater through, for example, inadvertent spillage or leakage from petroleum fuel tanks (Corseuil and Alvarez, 1996).

Treatment of toluene-containing groundwater and wastewater has been studied by many authors using a variety of bioreactor types under both aerobic and anaerobic conditions. For example, Ahmadvand et al. (1995) achieved 99% removal efficiency by aerobically treating BTEX-contaminated wastewater (14 mg toluene l^{-1}) using fluidized bed reactors. De Nardi et al. (2002, 2005) indicated the viability of using anaerobic horizontal-flow anaerobic immobilized biomass (HAIB) reactors to treat wastewater and ground water contaminated with BTEX and petroleum products.

Consequently, the literature suggests that anaerobic digestion (AD) can and should be fully exploited as a remediation

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option for toluene-contaminated wastewaters. The treatment of volatile compounds, such as toluene, by AD has several advantages over aerobic treatment. Firstly, losses to the atmosphere due to agitation and aeration of the wastewaters are avoided. Secondly, in the absence of these devices/requirements the energy input required by a treatment plant is considerably reduced. Moreover, anaerobic systems offer the added benefit of methanogenic biogas production, resulting in a renewable energy source (>65% methane), which can be used for the generation of heat and electricity for both industrial and domestic use (McHugh et al., 2003). These advantages, coupled with significantly reduced excess sludge production presents AD as an attractive alternative wastewater treatment solution.

In addition, and as many wastewaters are released for treatment at (sub)-ambient temperatures (Lettinga et al., 2001), low-temperature, or psychrophilic, anaerobic digestion (PAD) can present a more attractive and cost-effective wastewater treatment option. Specifically, PAD can greatly reduce losses incurred by wastewater/bioreactor heating, which can consume up to 30% of biogas produced (Collins et al., 2005b). Indeed, the feasibility of PAD has been previously demonstrated for a wide range of wastewater categories (Collins et al., 2003, 2004; McHugh et al., 2004, 2005), including toxic and carcinogenic compounds of serious concern with respect to environmental protection and human health, such as trichlorophenols (TCP) (Collins et al., 2005b).

In the context of the above discussion, the objective of this study was to assess the feasibility of low-temperature (15 °C) AD of toluene-containing, synthetic industrial wastewaters using the expanded granular sludge bed bioreactor design.

2. Materials and methods

2.1. Source of biomass

A mesophilic anaerobic sludge was obtained from a full-scale (1500 m^3) internal circulation (IC) bioreactor, operated at 37 °C for the treatment of citric acid production wastewater, at the Archer Daniels Midland (ADM) citric acid production plant, Ringaskiddy, Co. Cork, Ireland . The volatile suspended solids (VSS) concentration of the sludge, which consisted of well-settling brown and grey spherical granules (1–2 mm), was 60.5 $g\,l^{-1}$.

2.2. Design and operation of EGSB bioreactors

Two glass, laboratory-scale (3.52 l), expanded granular sludge bed-anaerobic filter (EGSB-AF) reactors, R1 and R2, which were of the same design as those described by Collins et al. (2005a), were each inoculated with 60 g VSS of granular sludge. R1 and R2 were used for the stabilization of a volatile fatty acid (VFA)-based, synthetic wastewater consisting of ethanol, butyrate, propionate and acetate, in the chemical oxygen demand (COD) ratio of 1:1:1:1, to a total of 5 g COD l^{-1}. The influent was buffered with $NaHCO_3$ and fortified, as described by Shelton and Tiedje (1984), with macro- (10 ml l^{-1}) and micro- (1 ml l^{-1}) nutrients. Both reactors were maintained at 15 °C throughout the 630-d trial period. The organic loading rate (OLR) applied to R1 and R2 was 2.8 kg COD $m^{-3}\,d^{-1}$, with a hydraulic retention time (HRT) of 48 h and a sludge loading rate (SLR) of 0.028 $m^3_{wastewater}$ kg $[VSS]^{-1}\,d^{-1}$. Effluent was re-circulated through the systems at an applied upflow velocity of 5 m h^{-1}. The applied HRT was reduced to 24 h on day 55, thus increasing the applied OLR to 5.7 kg COD $m^{-3}\,d^{-1}$. R2 influent was supplemented with toluene (solubilised in ethanol) on day 300, to a final concentration of 5 mg l^{-1}, to give a toluene loading rate (TLR) of 1.42 g toluene $m^{-3}\,d^{-1}$. Toluene was not added to the R1 influent wastewater, thereby maintaining this reactor as an experimental control. The trial period was divided into eight operational phases (P1–P8, as presented in Table 1).

2.3. Analytical methods

Biogas and effluent from R1 and R2 were routinely sampled and temporal biogas methane content and effluent COD concentrations were determined according to Standard Methods (APHA, 1998). Effluent toluene concentrations (R2 only) were ascertained using high performance liquid chromatography (HPLC) analysis (liquid chromatograph LC-6A, UV spectrophotometric detector SPD-6A and chromatopac C-R1A; Shimadzu, Japan), whereby samples were separated on a hypersil-ODS C18 column (2.1 mm i.d. × 100 mm long × 3 μm particle size) using a mobile phase containing acetonitrile: H_2O (70:30), a detection wavelength of 254 nm and flow rate of 1.5 ml min^{-1}.

2.4. Specific methanogenic activity (SMA) and toxicity testing

The metabolic status of the seed biomass, and samples collected from the bioreactors on day 250 and 630 (trial conclusion), was assessed using SMA assays, which were performed using the pressure transducer technique (Colleran et al., 1992; Coates et al., 1996). Acetate and H_2/CO_2 were employed as substrates, in order to establish the activity of acetoclastic and hydrogenotrophic methanogens, respectively, and propionate, butyrate and ethanol were used to assay indirect methanogenic metabolism coupled to syntrophic activity. SMA assays were also performed using biomass removed from the fixed-film section of the bioreactors at the conclusion of the trial.

Toluene methanogenic toxicity of the seed and the reactor (R2) biomass was assessed using SMA-based toxicity assays (acetoclastic and hydrogenotrophic) as described by Colleran and Pistilli (1994) and Enright et al. (2005). Toxicity was defined in terms of the IC_{50} value i.e., the concentration (mg l^{-1}) of toluene that resulted in 50% inhibition of SMA, which was calculated from the linear regression of SMA as a function of toluene concentration. It should be noted, however, that, due to the lack of acetoclastic activity, toxicity tests were carried out only with respect to the hydrogenotrophic community at the conclusion of the R2 reactor trial. All activity and toxicity batch assays contained 2–5 g VSS l^{-1} and were performed at 15 and 37 °C (psychrophilic and mesophilic conditions, respectively).