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Research article

Assessing the treatment of acetaminophen-contaminated brewery wastewater by an anaerobic packed-bed reactor

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A B S T R A C T

The treatment of high-strength organic brewery wastewater with added acetaminophen (AAP) by an anaerobic digester was investigated. An anaerobic packed-bed reactor (APBR) was operated as a continuous process with an organic loading rate of 1.5-g COD per litre per day and a hydraulic retention time of three days. The results of steady-state analysis showed that the greatest APBR performances for removing COD and TOC were as high as 98 and 93%, respectively, even though the anaerobic digestibility after adding the different AAP concentrations of 5, 10 and 15 mg L^{-1} into brewery wastewater can affect the efficiency of organic matter removal. The average CH_4 production decreased from 81 to 72% is counterbalanced by the increased CO_2 production from 11 to 20% before and after the injection of AAP, respectively. The empirical kinetic models for substrate utilisation and CH_4 production were used to predict that, under unfavourable conditions, the performance of the APBR treatment process is able to remove COD with an efficiency of only 6.8%.

1. Introduction

Pharmaceutical industry wastewater may contain high-value active ingredients, such as acetaminophen, which is classified as recalcitrant because of its physiological storage behaviour. Pharmaceutically active compounds (PACs) present in wastewater can affect the biodiversity and ecology of the receiving waters when released without treatment (Jones et al., 2004; Mendoza et al., 2015). The occurrence and persistence of PACs and their metabolites have been detected in sewage treatment plant effluents, surface waters, and, less frequently, in ground water and drinking water even though the selection and control of a safe and effective therapeutic dose in human and veterinary practices can be made (Sun et al., 2014; Tambosi et al., 2010). The bacterial toxicity of PACs may play an important role in decreasing the performance of biodegradable organic matter removal in affected treatment systems (Sponza and Demirden, 2007). Acetaminophen, or paracetamol (N-acetyl-4-aminophenol) (AAP), is the most frequently used analgesic and antipyretic drug (Narang et al., 2015). In the European Union, sewage treatment effluents were identified as the point source of active ingredients in the river water, with AAP concentrations up to 6 μg L^{-1}, even though more than 65 μg L^{-1} of AAP was reported in the Tyne River, UK (Duran et al., 2011). The presence of PACs, such as the analgesic AAP in natural waters, has been detected at concentrations of 10 μg L^{-1} in the USA (Kolpin et al., 2002). PACs can be classified as newly emerging pollutants (NEPs) (Bell et al., 2011; Murray et al., 2010; Zenker et al., 2014) and may cause subtle effects on aquatic and terrestrial organisms due to their virtual ubiquity in various environments (Kummerer, 2011; Zuccato et al., 2006). Among the major categories of NEPs are pharmaceuticals and illicit drugs, steroid oestrogens (hormones and contraceptives) and personal care products (Mostafa et al., 1990; Nikolaou, 2013; Ramachandran and Saraswathy, 2014). Therefore, the effectiveness of biological and physical treatment

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processes in the removal of pharmaceuticals and other organic compounds needs to be verified (Galhetas et al., 2014; Stackelberg et al., 2007; Yoon and Byun, 2013; Zaib et al., 2013).

Wastewaters of pharmaceutical industries generally contain high organic loads, and treatment is primarily conducted using two major types of aerobic and anaerobic digestion (Novak et al., 2003), even though it can be advanced by an alternating anoxic aerobic process to remove inorganic nitrogen pollution (Fulazzaky et al., 2015). Anaerobic digestion processes have been widely used for the treatment of high-strength industrial wastewaters containing herbicides, antibiotics, phenols, cosmetics, etc. (Abdullah et al., 2013; Chen et al., 2011). A hybrid bioreactor of hollow fibre microfilter membrane and cross-linked enzyme aggregates has been used for the elimination of AAP (Ba et al., 2014). Consequently, in this work, an anaerobic packed-bed reactor (APBR) is used to treat AAP-contaminated brewery wastewater due to the large packing media surface area available for the attachment of microorganisms and the reduced bioreaction time (Jong and Parry, 2003). The use of a membrane or combined membrane bioreactor could be useful for the treatment of synthetic pharmaceutical wastewater containing AAP or trace organic contaminants (Nguyen et al., 2013; Shariati et al., 2010). Cascaded anaerobic ponds are the most commonly used process for the treatment of wastewaters to withstand high organic loading rates, such as for palm oil mill effluent (Fulazzaky, 2013). High-rate anaerobic treatment of pharmaceutical wastewaters in a packed-bed biofilm reactor with the various types of supporting materials possesses a basic understanding of fixed-film biological reactor processes (Gullicks et al., 2011; Satya and Venkateswarlu, 2013). The materials used to retain active biomass in the reactor can be arranged in various confirmations made out of different materials, such as plastics, granular activated carbon, sand reticulated foam polymers, granite, quartz and stones, and can be loosely or modularly packed. The advantages of using the supporting materials as biofilm carriers are that they can assure a shorter start-up period and a greater amount of retained inoculum for faster start-up (Kim et al., 2004). APBR would be suitable for the treatment of high-strength wastewaters and has the traditional biofilm resistance to shock loading and biological inhibition (Scullion et al., 2007); this can be operated in either an up-flow or down-flow feed mode (Nandy and Kaul, 2001; Yu and Gu, 1996). Although the treatment of pharmaceutical wastewaters containing synthetic drugs by anaerobic digesters has been widely studied in the last two decades (de Graaff et al., 2011; Lin et al., 2012; Masse et al., 2000), the kinetic models of substrate utilisation and methane (CH_4) production in treating the AAP-contaminated wastewater by an APBR needs to be established to predict digester performance under unfavourable conditions. This may contribute to a better understanding of the application and effectiveness of the anaerobic digestion process for the removal of PACs from wastewaters.

The objectives of this study are as follows: (1) to assess the performance of APBR for the treatment of AAP-contaminated brewery wastewaters, (2) to monitor the fluxes of CH_4 and carbon dioxide (CO_2) from a small-scale anaerobic digester to the atmosphere, and (3) to investigate the kinetics of substrate utilisation and CH_4 production in an anaerobic digestion process for treating a high-strength organic wastewater under mesophilic temperature conditions.

2. Materials and methods

2.1. Anaerobic packed-bed reactor

The APBR treatment system (dos Reis and Silva, 2014; Ferraz et al., 2014; Singh and Prerna, 2009) used in this study consists of a 22.5-L cylindrical PVC bioreactor, 10-L raw wastewater storage tank, 0.5-L Schott bottle filled with AAP and 10-L effluent tank, as shown in Fig. 1. The bioreactor filled with plastic-based packing media can treat the wastewater in an up-flow feed mode. The fragmented pieces of polyurethane pipe, with an inside diameter of 0.64 cm, outside diameter of 0.95 cm and density of 900 kg m^{-3}, were used as a matrix to immobilise microorganisms. The percentage of void space in the bioreactor was approximately 85%, with an effective volume of 18.5 L. The addition of AAP from the Schott bottle was regulated using a 230-V centrifugal water pump (Totton Pump Limited, Southampton, England) to allow the joining of it to the raw wastewater feeding the APBR. The fed wastewater enters the APBR treatment system through a downcomer tube of 0.6 L inside the cylindrical PVC bioreactor. The experimental set up used during this study was maintained at mesophilic conditions (37 °C), and thus the bioreactor sidewall was enclosed within the tubular PVC water-jacket connected to a heat exchanger. The circulations of fed wastewater and effluent were regulated using the centrifugal pumps, and thus the upflow velocity was 1.25 cm h^{-1}. The gas pipeline that has access to biogas production in the bioreactor was connected to an optical gas bubble counter (made in-house at Newcastle University, UK), giving a measurement of the gas volume. The biogas production was monitored and collected in the range of 0–1.5 L h^{-1}. The APBR treatment system was equipped with two sampling ports that allowed biological solids and liquid samples to be withdrawn periodically for quantitative analysis throughout the experiment.

2.2. Operating procedures

One litre of anaerobic granular sludge from an anaerobic sludge digester at the municipal wastewater treatment plant of Hexham town, Northumberland, UK, was used as the microbial inoculum for the start-up of the APBR treatment process. The brewery wastewaters (Scottish and Newcastle Breweries, Newcastle, UK) have an average chemical oxygen demand (COD) concentration of 88 g L^{-1}; therefore, the wastewater used was diluted to 5.68% of the original concentration with potable water to have an influent COD concentration of 5 g L^{-1}. The initial average mixed liquor volatile suspended solids (MLVSS) concentration in the APBR was appoximately 6 g L^{-1}. To assess the effect of AAP on the APBR performance, the Schott bottle was filled with the AAP of Calpol Six Plus 250-mg/5-mL Suspension. The inlet arrangement in the wastewater-fed APBR used a centrifugal pump control to achieve a desired concentration of AAP in the diluted brewery wastewaters. The anaerobic digestion system was operated at 37 °C with an organic loading rate (OLR) of 1.5-g COD per L per day and a

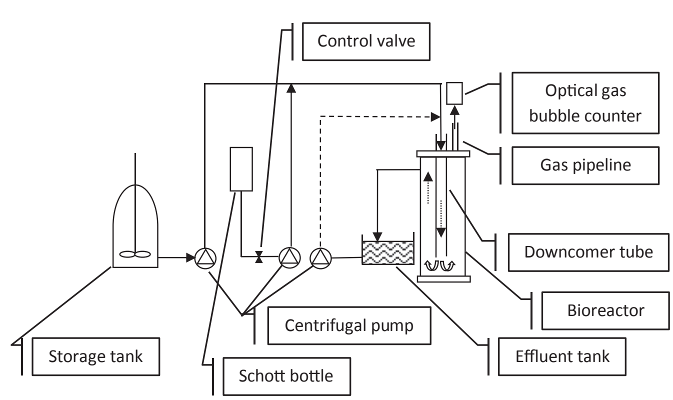

Fig. 1. Schematic of the anaerobic packed-bed reactor.