Bioresource Technology 236 (2017) 1–10

Contents lists available at ScienceDirect

Bioresource Technology

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

Performance and microbial community structure in an integrated anaerobic fluidized-bed membrane bioreactor treating synthetic benzothiazole contaminated wastewater

Yue Li [a], Qi Hu [a,c], Chun-Hong Chen [a], Xiao-Long Wang [a], Da-Wen Gao [a,b,*]

[a] State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China
[b] School of Forestry, Northeast Forestry University, Harbin 150040, China
[c] School of Pharmaceutical Engineering, Shenyang Pharmaceutical University, Shenyang 110016, China

HIGHLIGHTS

- Integrated anaerobic membrane bioreactor was used to treat benzothiazole wastewater.
- The accumulation of VFAs increased with the addition of benzothiazole.
- The biodegradation of benzothiazole was increased by the adaptation of microbes.
- Acetotrophic methanogens were more sensitive to the addition of benzothiazole.

ARTICLE INFO

Article history:
Received 31 January 2017
Received in revised form 29 March 2017
Accepted 30 March 2017
Available online 2 April 2017

Keywords:
Integrated anaerobic fluidized-bed membrane bioreactor
Benzothiazole
Anaerobic process
Biodegradation kinetics
Microbial community structure

ABSTRACT

This study investigated the impact of benzothiazole on the performance and microbial community structures in an integrated anaerobic fluidized-bed membrane bioreactor fed with synthetic benzothiazole wastewater (with gradually increasing doses of benzothiazole (1–50 mg/L)). The addition of benzothiazole had an adverse effect on volatile fatty acids accumulation (from 10.86 mg/L to 57.83 mg/L), and membrane fouling (service period from 5.9 d to 5.3 d). The removal efficiency of benzothiazole was 96.0%. Biodegradation was the major benzothiazole removal route and the biodegradation efficiency obviously improved from 25.7% to 98.3% after adaptation. Sludge 1 (collected on day 58 without benzothiazole) and sludge 2 (collected on day 185 with 50 mg/L benzothiazole) were analyzed using the Illumina®MiSeq platform. The most abundant genera were *Trichococcus* (43.1% in sludge 1) and *Clostridium sensu stricto* (23.9% in sludge 2). The dominant genus of archaea was *Methanosaeta* (90.3% in sludge 1 and 80.8% in sludge 2).

1. Introduction

Benzothiazole (BTH) and benzothiazole derivatives (BTHs) belong to the group of polar heterocyclic contaminants that contain a benzene ring and a thiazole-ring (El-Bassi et al., 2010). Due to widespread use in some industries (such as fungicides in lumber and leather production) (Reemtsma et al., 1995) and pharmacies (such as chemotherapeutic agents) (Bujdáková et al., 1993), BTH and BTHs have been detected in surface water (Loos et al., 2009; Ni et al., 2008). The limited biodegradability, the potential danger of mutagenic effects, and the allergenicity of BTHs make their presence in the environment as a great concern (El-Bassi et al., 2010). Studies have concentrated on the biodegradation rate of BTHs by aerobic condition (De Wever et al., 1998), pure culture bacteria (Chorao et al., 2009) and electro-assisted microbial reactor (Liu et al., 2014). However, aerobic process has higher energy costs compared to anaerobic treatment (Lew et al., 2009).

Wastewater treated by anaerobic process has been regarded more as a resource rather than a waste (McCarty et al., 2011; Wang et al., 2012). Due to the production of methane which can generate electricity and can be used as fuel, anaerobic process are widely used in wastewater treatment. Meng et al. treated diluting antibiotic fermented liquids by anaerobic EGSB reactor (Meng et al., 2015). They found that 85% and 80% removal of COD and amoxicillin were achieved. Furthermore, anaerobic process has been successful applied for the treatment of industrial wastewater

* Corresponding author at: State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China.
E-mail address: gaodw@hit.edu.cn (D.-W. Gao).

http://dx.doi.org/10.1016/j.biortech.2017.03.189
0960-8524/

at full scale (Akarsubasi et al., 2006; Niwa et al., 2016), including treatment of antibiotic contaminated wastewater (Chen et al., 2011).

The combination of anaerobic and membrane reactors could improve the effluent quality (Dutta et al., 2014; Gao et al., 2014a; Qiu et al., 2013a). Moreover, the existence of membrane is a barrier for microbe biomass, resulting in increased activity (Tao et al., 2012). Some researchers investigated the anaerobic membrane bioreactor treating pharmaceutical wastewater (Svojitka et al., 2017). They found that the COD efficiency was obviously affected by varying wastewater composition. In addition, the Membrane Biological Reactor has demonstrated a relatively low release of antibiotic-resistant bacteria and antibiotic-resistant genes compared to the performance of four other types of wastewater treatment utilities (Activated Sludge, Oxidative Ditch, Rotatory Biological Contactors and Multiple Sludge treatment processes) (Munir et al., 2011). But, little information is available about the impact of BTH on the performance and microbial structure in an integrated anaerobic fluidized-bed membrane bioreactor.

This study investigated the feasibility of an integrated anaerobic fluidized-bed membrane bioreactor (IAFMBR) to treat high-strength wastewater containing BTH. The main purposes of this study were to research the impact of BTH on the performance of the IAFMBR and on the succession of microbial community structures. Batch experiments were used to determine the main elimination pathway of BTH and the biodegradation kinetics.

2. Materials and methods

2.1. IAFMBR and operation

The design of the IAFMBR referenced the previous design (Gao et al., 2014b). The configuration of the reactor was in Fig. S1. The reactor was made of 10 mm Plexiglas® and the total volume was 8.9 L (effective volume 6.1 L). The reactor was composed of an outer tube, a middle tube and an inner tube. The internal diameter and height of the outer, middle and inner tubes are 120 mm × 450 mm, 70 mm × 582 mm and 40 mm × 620 mm, respectively. Furthermore, the inner zone was equipped with a hollow fiber membrane (Mitsubishi Rayon Co., Ltd. Tokyo, Japan) that had a total surface membrane area of 0.21 m^2 and a pore diameter of 0.4 μm. The designed membrane flux was 11.3 $L/m^2{\cdot}h$.

The IAFMBR consisted of an AFBR (anaerobic fluidized-bed reactor) and an AnMBR (anaerobic membrane bioreactor). The treatment performance was investigated by the effluent of AFBR and IAFMBR. The AFBR effluent was treated by anaerobic granular sludge. The IAFMBR effluent was treated not only by anaerobic granular sludge, but also by the AnMBR.

The operation of the IAFMBR can be divided into three phases. The first phase was a start-up phase (phase I) of 58 days (1–58 d) during which there was no BTH in the influent. The second phase was the adaptation phase (phase II) of 93 days (59–151 d) during which the influent included gradually increasing concentrations of BTH (from 1 mg/L to 50 mg/L). The last phase was the stable adaptation phase (phase III) of 34 days (152–185 d) during which the influent BTH concentration was 50 mg/L. The influent flow was 6.1 L/d and the recycle ratio was at 35. The membrane effluent was continuous (no relaxation). During all stages, the IAFMBR was maintained at a hydraulic residence time of 24 h and temperature of 35 °C.

2.2. Inoculation and feed composition

The reactor was inoculated with anaerobic granular sludge taken from an anaerobic reactor in DaQing, China treating wastewater from alcohol production. A total of 1.2 L of sludge was added to the reactor, and the final mixed liquor volatile suspended solids concentration was 4850 mg/L.

The reactor was fed with synthetic wastewater containing BTH according to the characteristics of antibiotic production wastewater coming from a pharmaceutical factory in Harbin, China. The concentration of BTH from this pharmaceutical factory was about 50 mg/L. The main carbon sources were glucose and acetate to maintain the COD concentration at 3000 mg/L. The inorganic nutrient composed according to the previous study (Gao et al., 2014b).

Benzothiazole (CAS no. 95-16-9, C_7H_5NS, MW 135.19) was purchased from Aladdin Industry Corporation. To prevent acidification, sodium bicarbonate was used as the pH buffering agent.

2.3. Batch experiments and biodegradation kinetics models

To determine the main degradation pathway of BTH, batch experiments were conducted (Table 1). Two sludge samples (sludge 1 and sludge 2) were collected from the AFBR reactor on day 58 (the end of the start-up phase) and day 185 (the end of the stable adaptation phase) at different samling points, and then mixed them. Sludge 1 was not exposed to BTH.

In this test, 200 mL anaerobic bottles containing 100 mL mixed liquor were incubated at 35 °C for 84 h following the treatments (B, C, B' and C') (Table 1). The mixed liquor suspended solids concentration was 3102 ± 170 mg/L. The treatment A and A' were no sludge addition, but 100 mL deionized water instead of 100 mL mixed liquor. All experiments were conducted in duplicate.

All batch experiments were conducted in a closed chamber to avoid possible photolysis, and the activity of the anaerobic granular sludge was inhibited by NaN_3 (Li and Zhang, 2010). The concentration of BTH was 50 mg/L. Samples of the mixed liquor were taken for analysis from the batch experiments at the following times: 0, 0.5, 1, 2, 5, 8, 12, 24, 36, 48, 60, 72, and 84 h.

Three biodegradation kinetics models were applied to describe the biodegradation data, namely zero-order, first order (Eq. (1)) and second-order models (Li and Zhang, 2010).

$$\frac{dC}{dt} = -k_1 \cdot C \Longleftrightarrow C_t = C_0 \cdot e^{-k1t} \quad (1)$$

In Eq. (1), C_0 is the initial concentration of the BTH; C_t is BTH concentration at time t; and k_1 is the first-order rate constant. Half-life ($t_{1/2}$) was calculated as Eq. (2).

$$t_{1/2} = \ln 2/k_1 \quad (2)$$

Table 1
Batch experimental design.

Treatment	Sludge[a]	Benzothiazole (50 mg/L)	0.2% NaN_3[b]	Removal routes[c]
A	–[e]	+[d]	+	H
B	Sludge 1	+	+	H + A
C	Sludge 1	+	–	H + A + B
A'	–	+	+	H
B'	Sludge 2	+	+	H + A
C'	Sludge 2	+	–	H + A + B

[a] Sludge was collected at the end of the start-up phase (day 58, sludge 1) and the end of the stable adaptation phase (day 185, sludge 2).
[b] NaN_3 was used to inhibit the sludge biodegradation activity.
[c] B-biodegradation; A-adsorption; H-hydrolysis.
[d] "+" indicates presence.
[e] "–" indicates absence.