Process Biochemistry 51 (2016) 399–407

Contents lists available at ScienceDirect

Process Biochemistry

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

Biodegradation of a high-strength wastewater containing a mixture of ammonium, aromatic compounds and salts with simultaneous nitritation in an aerobic granular reactor

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A R T I C L E I N F O

Article history:
Received 21 September 2015
Received in revised form 27 December 2015
Accepted 31 December 2015
Available online 6 January 2016

Keywords:
Partial nitrification
Phenolic compounds
Salinity
Nitrous oxide

A B S T R A C T

Long-term operation (390 days) of a continuous airlift reactor with aerobic granular biomass was successfully applied to treat a highly complex wastewater composed of: ammonium (1000 mg N L^{-1}), *o*-cresol (100 mg L^{-1}), phenol (100 mg L^{-1}), quinoline (50 mg L^{-1}) and salts (16 g salts L^{-1}). High nitrogen loading rate (1.1 g N L^{-1} d^{-1}) and organic loading rate of 0.7 (g COD L^{-1} d^{-1}) were achieved for the simultaneous nitritation and complete biodegradation of the aromatic compounds. The successful operation of the granular airlift reactor can be related to (i) the growth of specialized microorganisms in the aerobic granules and (ii) the continuous feeding regime. Aerobic granules were maintained stable in spite of the high salinity conditions. Dissolved oxygen (DO) concentration and DO/ammonium concentrations ratio were the key parameters to select a suitable effluent for anammox or heterotrophic denitrification via nitrite. Besides, nitrous oxide emissions were related to the DO concentration in the reactor.

1. Introduction

Nowadays, the satisfactory and cost-effective treatment of industrial wastewaters from chemical, petrochemical, coke plant and refineries is a huge challenge since they are complex matrices, containing several organic and inorganic compounds, such as toxic/recalcitrant organic compounds (like aromatic compounds), ammonium and several inorganic salts [1,2]. Industrial wastewaters are often treated by physico-chemical processes; however, these technologies have serious drawbacks [3]: (i) high costs due to the high temperature and pressure conditions applied and the chemicals required, (ii) do not allow complete degradation of the aromatic compounds and (iii) they may produce other hazardous by-products (secondary pollutants).

Biological processes can overcome some of the disadvantages of physico-chemical treatments. However, biological processes can be inhibited by aromatic compounds [4]. Biological treatments based on floccular biomass (such as activated sludge systems) are the main biological technology used at full-scale. Nevertheless, its application for treating complex industrial wastewaters is limited because activated sludge systems are known to be highly inhibited by aromatic compounds [2–5]. Biological systems with floccular biomass: (i) are poor adjustable to fluctuations in the organic loading, (ii) need large reactor volumes because low loading rates can be applied and (iii) its settling capacity is limited, which means large secondary clarifiers. Biological treatments based on aerobic granules represent an alternative to treat these complex industrial wastewaters [6].

Ammonium can be removed by the application of Biological Nitrogen Removal (BNR) processes via nitrite [7,8], where ammonium is oxidized until nitrite (nitritation) and subsequently, nitrite can be reduced by heterotrophic denitrification via nitrite (denitritation) or by anammox (anaerobic ammonium oxidation) processes [7,8]. Nitrification is usually the bottleneck for BNR in many wastewater treatment processes because nitrifying bacteria are very sensitive to several factors, such as inhibition by aromatic compounds. Biodegradation of aromatic compounds and nitritation can be simultaneously performed in a single aerobic granular reactor due to the substrates gradients occurring through the granules and the high biomass concentration that can be achieved in this kind of reactor [6,9]. Therefore, a two-stage process composed by a first aerobic granular reactor followed by a second anoxic granular reactor has been proposed as a feasible technology to treat complex industrial wastewaters [9,10]. Until now, this option has

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http://dx.doi.org/10.1016/j.procbio.2015.12.020
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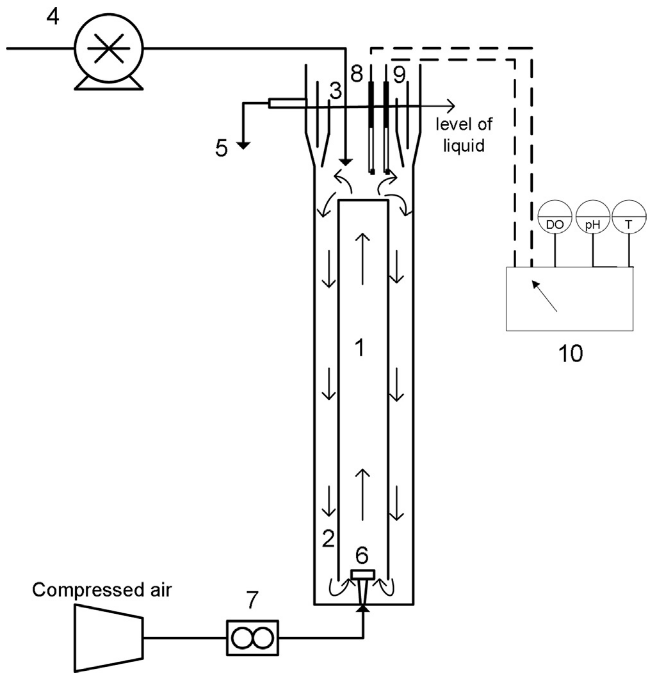

Fig. 1. Experimental set-up of the continuous granular airlift reactor. (1) riser; (2) down-comer; (3) separator; (4) feed pump; (5) effluent port; (6) air sparger; (7) rotameter; (8) pH probe; (9) DO probe; (10) monitoring panel.

only been tested with wastewaters containing ammonium and a single aromatic compound: *o*-cresol [9] or *p*-nitrophenol [10]. However, it is well-known that industrial wastewaters can be composed by a mixture of several aromatic compounds; thus nitritation and biodegradation of a mixture of aromatic compounds should be tested to confirm the technological feasibility of this option. Furthermore, the effect of several parameters should be established to guarantee this feasibility: (i) operation at high salinity conditions, (ii) determination of nitrous oxide emissions from nitritation at different dissolved oxygen (DO) concentrations in the reactor and (iii) assessment of the stability of the morphological characteristics of the aerobic granules at long-term. Therefore, the objective of this study is to demonstrate the feasibility of the biodegradation of a high-strength wastewater containing a mixture of ammonium, aromatic compounds and salts with simultaneous nitritation and aromatic compounds degradation in an aerobic granular reactor. In this sense, nitrous oxide emissions, stability and characteristics of the aerobic granular biomass at long-term operation and the microbial diversity in the granules will be assessed.

2. Materials and methods

2.1. Reactor

An airlift reactor made of glass (2.6 L of working volume) was used (Fig. 1). The reactor configuration was as follows: the internal diameter of down-comer was 62.5 mm; the riser had a height of 750 mm and an internal diameter of 42.5 mm and it was at 8 mm from the bottom of the down-comer. At the top of the reactor, a gas–solid–liquid separator allowed the separation of the aerobic granules from the treated wastewater. Compressed air was supplied through an air diffuser placed at the bottom of the reactor at an upflow velocity of 0.2–0.3 cm s^{-1}. Air flow rate in the reactor was regulated manually between 150 and 250 mL min^{-1} by a rotameter (Aalborg, USA) and it was enough to ensure an appropriate flow in the airlift reactor. The reactor was equipped with DO (Crison DO 6050), temperature (Crison Pt1000) and pH probes (Crison pH 5333) that were connected to a data monitoring system (Crison Multimeter 44). DO was not automatically controlled and varied between 0.5 and 4.0 mg O_2 L^{-1} (excluding the first 25 days) according to the applied airflow (Fig. 2A). A Programmable Logic Controller (PLC) coupled to a Supervisory Control And Data Acquisition (SCADA) system regulated temperature, pH and feeding. pH was maintained at 8.0 ± 0.2 by a regular addition of $NaHCO_3$ whereas temperature in the reactor was maintained at 30 ± 0.5 °C using a temperature controller coupled with a belt-type heating device (Horst, Germany). Feeding to the reactor was made with a membrane pump (ProMinent Gamma/L).

2.2. Inoculum

Aerobic granular sludge from an airlift reactor performing partial nitritation and *o*-cresol biodegradation was used as inoculum. The biomass characteristics were as follows: 1.0–1.5 mm of mean granule size, 40–60 m h^{-1} (67–100 cm min^{-1}) of settling velocity, sludge volumetric index at 5 min (SVI_5) of 7–14 mL g^{-1} TSS and SVI_{30}/SVI_5 ratio of 1.0. The biomass was under starvation period in mineral medium during 30 days at room temperature prior to the start-up of the reactor. More information can be found in Jemaat et al [9].

2.3. Wastewater composition and operational conditions

The airlift reactor was fed continuously with a synthetic wastewater. Throughout the operational period, ammonium concentration was of 1000 ± 39 mg N–NH_4^+ L^{-1} (3818 mg NH_4Cl L^{-1}) in the influent and different aromatic compounds were also added. During the first 60 days of operation, hydraulic residence time (HRT) was varied between 6.3 to 0.9 days (by increasing the inflow rate) to increase the applied organic (OLR) and nitrogen (NLR) loading rates. From day-60 onwards, HRT was maintained at 0.9 days. The complexity of the treated wastewater was increased stepwise by introducing the aromatic compounds as follows:

i) Phase I (from day-22 to day-114). Ammonium (1000 ± 39 mg N L^{-1}) and *o*-cresol (100 ± 6 mg L^{-1}) were simultaneously treated.
ii) Phase II (from day-114 to day-195). Phenol was added to the influent; therefore ammonium (1000 ± 39 mg N L^{-1}), *o*-cresol (100 ± 6 mg L^{-1}) and phenol were treated together. Phenol concentration in the influent was increased stepwise from 25 ± 1 to 100 ± 3 mg L^{-1}.
iii) Phase III (from day-195 to day-332). Quinoline was added to the influent; therefore ammonium (1000 ± 39 mg N L^{-1}), *o*-cresol (100 ± 6 mg L^{-1}), phenol (100 ± 3 mg L^{-1}) and quinoline were treated together. Quinoline concentration in the influent was increased stepwise from 15 ± 2 to 50 ± 4 mg L^{-1}.

The aim of the first three phases was to achieve an effluent containing only nitrite, removing completely *o*-cresol, phenol and quinoline for a subsequent denitritation stage.

iv) Phase IV (from day-333 to day-390). The influent was the same than in phase III but the aim was to achieve an effluent with a nitrite/ammonium ratio close to 1 for a subsequent anammox stage, removing completely *o*-cresol, phenol and quinoline.

The chemical oxygen demand to nitrogen ratio (COD/N) in the influent increased from 0.25 to 0.64 throughout the experimental period. The composition of the micronutrients in the synthetic wastewater was (expressed as mg L^{-1}): 48 CH_3COONa; 12.5 $C_6H_{12}O_6$; 11.9 $C_{12}H_{22}O_{11}$; 41.0 KH_2PO_4; 176.0 NaCl; 198.0 MgCl2 × 7H2O; 4.0 $FeSO_4 \times 7H_2O$; 3.0 $MnSO_4xH_2O$; 4.0