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Long-term performance and stability of a continuous granular airlift reactor treating a high-strength wastewater containing a mixture of aromatic compounds

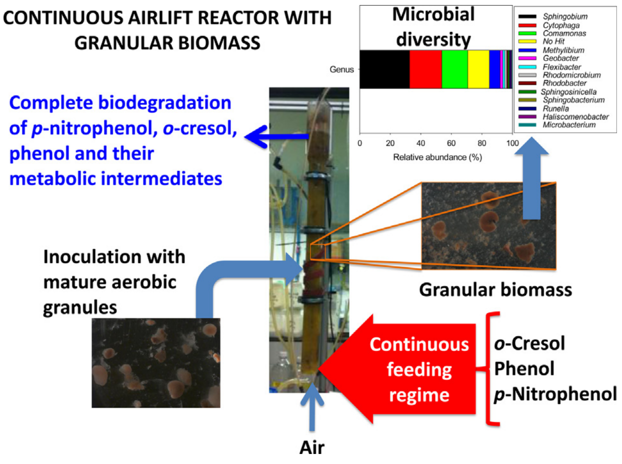
Carlos Ramos, María Eugenia Suárez-Ojeda, Julián Carrera*

GENOCOV Research Group, Department of Chemical, Biological and Environmental Engineering, School of Engineering, Universitat Autònoma de Barcelona, Edifici Q, 08193 Bellaterra, Barcelona, Spain

HIGHLIGHTS

- Aerobic biodegradation of a mixture of aromatics is feasible in a granular reactor.
- Applied organic loading rate is a key parameter for an optimal reactor performance.
- Stable mature aerobic granules were maintained 400 days in a continuous reactor.
- *Sphingobium*, *Cytophaga* and *Comamonas* were the main genera in the aerobic granules.

GRAPHICAL ABSTRACT



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ABSTRACT

Continuous feeding operation of an airlift reactor and its inoculation with mature aerobic granules allowed the successful treatment of a mixture of aromatic compounds (*p*-nitrophenol, *o*-cresol and phenol). Complete biodegradation of *p*-nitrophenol, *o*-cresol, phenol and their metabolic intermediates was achieved at an organic loading rate of 0.61 g COD L^{-1} d^{-1}. Stable granulation was obtained throughout the long-term operation (400 days) achieving an average granule size of 2.0 ± 1 mm and a sludge volumetric index of 26 ± 1 mL g^{-1} TSS. The identified genera in the aerobic granular biomass were heterotrophic bacteria able to consume aromatic compounds. Therefore, the continuous feeding regimen and the exposure of aerobic granules to a mixture of aromatic compounds make possible to obtain good granulation and high removal efficiency.

1. Introduction

Industrial wastewaters from agro-industries, coking plants, petrochemicals, pharmaceuticals, oil refineries, coal gasification processes, disinfectant/pesticides/fungicides and chemicals

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* Corresponding author.
E-mail address: julian.carrera@uab.cat (J. Carrera).

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manufacturing are complex matrices composed by several recalcitrant/toxic compounds, such as aromatic compounds [1,2]. Industrial wastewaters are often treated by physico-chemical processes. However, these technologies have serious drawbacks [3,4]: (i) high costs due to the required conditions of temperature and pressure and the use of some chemicals, (ii) incomplete degradation of the recalcitrant/toxic organic compounds and (iii) generation other hazardous by-products (secondary pollutants).

Biological processes can satisfactorily overcome some of the disadvantages of physico-chemical processes. Technologies based on flocculent biomass, such as activated sludge systems, are the main biological processes implemented at full-scale, however its practical application for treating complex industrial wastewaters is rather limited because activated sludge systems are known to be inhibited by aromatic compounds [4]. To overcome the inhibition caused by organic compounds, a promising alternative to activated sludge systems is the application of reactors with aerobic granular biomass [5]. The application of aerobic granules allows retaining slow growing microorganisms and protects them from high concentrations of pollutants due to the diffusion gradients generated along the granule [5] that favour the gradual adaptation of microorganisms to stressing conditions.

In the past, several studies showed that aerobic granules can be used to treat single aromatic compounds in sequencing batch reactors (SBRs), for example: phenol [6–8], nitrophenols [9–12], chlorophenols [13–15] and cresols [16]. In most of these studies, a readily biodegradable organic compound was used as co-substrate for biomass growth. However, to the best of the authors' knowledge aerobic granular-based technologies have been hardly applied to treat more complex wastewaters composed by a mixture of aromatic compounds. This could be related to several facts: (i) the granular biomass is mostly applied in SBRs, thus it can be inhibited because high concentrations of aromatic compounds can be accumulated at the beginning of the SBR cycle; this inhibition can be increased since the accumulation of several aromatic compounds can produce synergistic effects and (ii) the formation of aerobic granules from floccular biomass is not an easy task when some specific aromatics such as nitrophenols or chlorophenols are treated. For instance, Fernández et al. [9] and Suja et al. [10] failed to reach a stable aerobic granular SBRs at long-term treating aromatic compounds since *p*-nitrophenol (PNP) seemed to inhibit many microbial species involved in granulation [10,17]. Therefore, new strategies should be applied to achieve successful treatment of wastewaters containing mixtures of aromatics compounds with aerobic granular reactors. An alternative way to develop aerobic granules to treat aromatic compounds can be the inoculation with aerobic granules treating readily biodegradable substrates and its adaptation to consume aromatic compounds [8,10,13,18]. Furthermore, the use of a continuous reactor, such as an airlift reactor, instead of a SBR allows preventing inhibitory effects caused by the aromatic compounds and enhancing the granulation process.

Therefore, the aim of this study is to demonstrate that a wastewater containing a mixture of aromatic compounds (*p*-nitrophenol, phenol and *o*-cresol) can be successfully treated by an aerobic granular-based technology provided that: (i) mature aerobic granules are used as inoculum and (ii) a continuous airlift reactor is chosen instead of SBR.

2. Materials and methods

2.1. Reactor

An airlift reactor made of glass (2.6 L of working volume) was used. 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. Compressed air was supplied through an air diffuser placed at the bottom of the reactor at an up-flow velocity of 0.12–0.24 cm s^{-1}. Airflow rate in the reactor was regulated manually between 100 and 200 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 dissolved oxygen (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 but varied between 4.0 and 5.0 mg O_2 L^{-1} according to the applied airflow rate. 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

Mature aerobic granules from an airlift performing simultaneous partial nitrification and biodegradation of *p*-nitrophenol (PNP) was used as inoculum [19]. The characteristics of the inoculated granular biomass were: 2.4 ± 0.6 mm of mean granule size, 68 ± 23 m h^{-1} of settling velocity, 10 ± 1 mL g^{-1} TSS (total suspended solids) of sludge volumetric index (SVI) at 30 min (SVI_{30}), an SVI_{30}/SVI_5 ratio of 1.0 and a biomass density of 12.5 ± 2 g VSS $L^{-1}{}_{particle}$. The inoculum was mainly composed by ammonia-oxidizing bacteria (AOB) and *p*-nitrophenol-degraders (basically *Acinetobacter* genus). The amount of inoculum used was the same as the reactor volume (2.6 L) with a concentration of 2.8 g VSS (volatile suspended solids) L^{-1}.

2.3. Wastewater composition and operational conditions

The airlift reactor was continuously fed with a high-strength synthetic wastewater composed by a mixture of aromatic compounds (*p*-nitrophenol, phenol and *o*-cresol) and a readily biodegradable mixture of glucose and sucrose. The operational period was divided in 3 phases, where the wastewater complexity was progressively increased. During phase I (the first 181 days of operation), *p*-nitrophenol was the only treated aromatic compound and its concentration in the influent was increased stepwise from 39 ± 1 mg PNP L^{-1} (63 mg COD L^{-1}) to 371 ± 10 mg PNP L^{-1} (598 mg COD L^{-1}). Therefore, the total organic matter concentration in the wastewater (including *p*-nitrophenol, glucose and sucrose) was increased from 121 ± 4 mg COD L^{-1} to 1161 ± 5 mg COD L^{-1}. In this phase, the hydraulic retention time (HRT) varied between 1.2 to 10.4 days. Phase II started on day-181, after the addition of phenol in the influent. During phase II, phenol concentration in the influent was increased (in one step) from 114 ± 1 mg phenol L^{-1} (336 mg COD L^{-1}) to 598 ± 2 mg phenol L^{-1} (1417 mg COD L^{-1}) whereas *p*-nitrophenol concentration remained at 371 ± 10 mg PNP L^{-1} (598 mg COD L^{-1}). Throughout phase II, the total concentration of organic matter in the influent increased from 1842 ± 6 mg COD L^{-1} to 2530 ± 2 mg COD L^{-1} and the HRT ranged from 1.2 to 1.8 days. In phase III (from day-260 to day-400), an *o*-cresol concentration of 103 ± 3 mg *o*-cresol L^{-1} (259 mg COD L^{-1}) was treated simultaneously with *p*-nitrophenol (371 ± 10 mg PNP L^{-1}) and phenol (598 ± 12 mg phenol L^{-1}). Therefore, the influent concentration was maintained in this phase at 3486 ± 10 mg COD L^{-1} and the HRT was increased stepwise from 1.8 to 5.7 days. During the whole operation, the average SRT in the airlift was 39 ± 11 d.