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Investigation of rapid granulation in SBRs treating aniline-rich wastewater with different aniline loading rates

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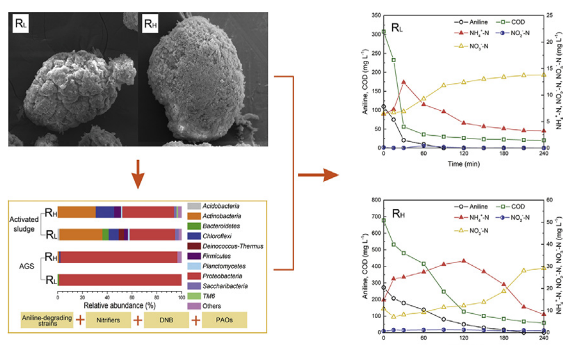
HIGHLIGHTS

- AGS was rapidly developed with different aniline loading rates (ALRs).
- High removal efficiencies of aniline, COD, and NH_4^+-N were achieved in the reactors.
- TN removal performance was better with higher concentration of aniline.
- *Proteobacteria* was dominant and *Actinobacteria* was enriched with higher ALRs.
- *Pseudomonas* plays a crucial role in aniline degradation as well as denitrification.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, aerobic granules were cultivated in two reactors which were denoted as R_L and R_H under 0.6 and 1.8 kg m^{-3} d^{-1} of aniline loading rates, respectively. The aerobic granular sludge (AGS) in the two sequential batch reactors for treating aniline-rich wastewater was compared. The results showed that the AGS could be rapidly formed with sludge volume index below 30 mL g^{-1}. The AGS in R_L had more filamentous bacteria than that in R_H by microstructural observations while the secretion of protein in extracellular polymeric substances was improved in R_H and in turn increased relative hydrophobicity of AGS. Within 4-h cycle, the excellent removal of aniline and chemical oxygen demand (COD) were achieved in the two reactors. The removal efficiencies of aniline and COD were consistently over 99.7%, 89.6%, respectively in R_L and 98.6%, 86.6%, respectively in R_H. As for nitrogen removal, NH_4^+-N released from aniline biodegradation could also be reduced efficiently via nitrification and no nitrite accumulation occurred in both the reactors. Total nitrogen removal performance in R_H was better, due to a more compact structure of AGS. The investigation of microbial community succession by pyrosequencing showed that the diversity of microorganisms decreased when AGS was developed. *Proteobacteria* especially *Gammaproteobacteria* significantly increased during aerobic granulation in both reactors. It was also found that the relative abundance of *Actinobacteria* was higher in R_H than that in R_L. Furthermore, the strains responsible for aniline biodegradation, nitrification, denitrification, and phosphorous accumulation were detected in the systems.

1. Introduction

Aniline is a key precursor and common intermediate in a variety of industries such as dyes, herbicides, pharmaceuticals, and rubber (Chen and Huang, 2015). It frequently appears in industrial effluents since it is widely used in industrial manufacturing and can be generated from

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biodegradation of xenobiotic compounds (Szczepanik and Słomkiewicz, 2016). It is detrimental to public health because of its carcinogenicity, and raises ecosystem imbalance due to its high toxicity to aquatic life. Considering the persistent nature of aniline and its wide-spreading globally, wastewater containing aniline requires proper treatment prior to discharge (Wen et al., 2015). Besides the conventional physical-chemical process, aniline-containing wastewater can also be treated via biodegradation. Through this process, aniline is firstly broken down to catechol and ammonia by aniline dioxygenase, and further the catechol could be degraded through cleavage of aromatic ring (Emtiazi et al., 2001). Since biological methods are cost-effective and environment-friendly, they are regarded as promising processes for wastewater treatment. However, aniline is toxic to many kinds of microorganisms, resulting in inhibition to their growth within the reactors (Vázquez and Rial, 2014). Furthermore, for many biological aniline removal systems, ammonia released from aniline biodegradation could be accumulated, which needs subsequent polishing steps for ammonia reduction (Dvořák et al., 2014; Wang et al., 2007).

Currently, aerobic granular sludge (AGS) technology as one of biological treatment processes is considered as an innovative strategy applied in treatment of both domestic and industrial wastewaters (Gao et al., 2011). AGS is developed based on the agglomeration of microbial consortia by a self-produced exopolymeric matrix (Henriet et al., 2016). Compared with activated sludge, AGS has compact structure with rapid settling property, resulting in higher biomass concentrations. Granulation also leads to the enrichment of slow-growing nitrifying bacteria, which is beneficial for enhanced nitrification activity in the system (Li et al., 2013). In addition, the AGS reactors can withstand shock loading and toxic conditions (Pijuan et al., 2009). For instance, AGS systems could maintain stable operation with 4 μM of shock loading provided by fluoxetine which was a chiral fluorinated pharmaceutical (Moreira et al., 2015) and could tolerate 32 μM shock of fluoroquinolones (Amorim et al., 2014). It has also been found that AGS reactors were able to degrade 250 mg L^{-1} of phenol (Wosman et al., 2016), 50 mg L^{-1} of *p* nitrophenol (Yarlagadda et al., 2012), and some other organic pollutants in wastewaters. As with cultivation of AGS for treating toxic compounds, for instance, it took 167 days to develop AGS capable of degrading 2,4 dichlorophenoxyacetic acid although half-mature aerobic granules acted as seed sludge (Quan et al., 2011). Also, in a sequential batch reactor (SBR) treating *o* nitrophenol-containing wastewater, stable aerobic granules were only formed after 122 days of operation (Basheer et al., 2012). During the last few years, reports regarding rapid granulation with toxic wastewater are rare especially without a large amount of easily biodegradable organics such as sodium acetate or glucose in the influent as co-carbon source. Furthermore, it has been found that the toxicity of these recalcitrant organics could even lead to the disintegration of granular structure under relative high loading conditions. In the report of Zhu et al. (2013), around 0.25 g L^{-1} d^{-1} of 4 chloroaniline loading could cause disintegration of AGS. When organic loading rate (OLR) was provided by refractory organic contaminants, metabolic intermediates accumulated in the reactors can be even more toxic than the original wastewater (Winkler et al., 2018). This suggested the importance to investigate the effect of loading rates of toxic recalcitrant organics on rapid aerobic granulation, especially to non-acclimatized seed activated sludge. Although a few reports have discussed AGS process with varying OLR (Liu and Tay, 2012; Peyong et al., 2012), to our knowledge, the effect of loading rates of toxic aromatic compounds in parallel reactors on rapid AGS development has yet to be documented.

In this work, rapid granulation was investigated in the reactors applied to treat aniline-containing influent with different aniline loading rates (ALRs). To shorten the start-up, aniline was directly used as sole source of carbon instead of replacement of common carbon source initially or step-wise increase of the concentration of the target pollutant. The properties of AGS in reactors fed with different concentrations of aniline were evaluated and compared. The removal efficiencies of aniline, chemical oxygen demand (COD) and nutrients during AGS development and further stable operation stages were determined. Furthermore, the abundance and structure of microbial consortia in both the AGS reactors were investigated by using pyrosequencing and the main contributors for granulation and pollutant reduction were further revealed. The present work assessed rapid granulation with the reactors directly fed with different concentrations of aniline, which would be significant for the practical application of the AGS technology in treating aniline-containing industrial effluents.

2. Material and methods

2.1. Experimental setup and operation conditions

Two identical lab-scale SBRs were used with 1 m in height and 0.08 m in diameter. The two reactors were denoted as R_L and R_H with regards to their low and high ALR conditions, respectively. The reactors were made of acrylic plastic and the effective working volume of the reactors was 4 L. Both reactors were inoculated with activated sludge collected from a biological tank of the Longwangzui municipal wastewater treatment plant located in Wuhan, China without previous exposure to aniline. The initial mixed liquor suspended solids (MLSS) concentrations of the two reactors were adjusted to 3500 ± 300 mg L^{-1}. A 4-h operation cycle was implemented by the automatic control of timers. Each cycle consisted of 5 min feeding, 220–225 min aeration, 5–10 min settling and 5 min decanting. When settling performance of the sludge improved noticeably, the settling phase of the operation cycle was gradually reduced from 10 min to 5 min to wash out the sludge with poor settleability and accelerate the granulation process. The aeration time was adjusted accordingly to maintain a constant cycle duration. A solenoid valve for effluent decant was located at the half height of the reactor to ensure a volume exchange ratio of 50%. During granule formation, the sludge retention time was not controlled to a certain level. The aeration was carried out by passing air through a porous stone diffuser at the bottom of the reactor using an air pump and the dissolved oxygen (DO) concentration in aeration phase was controlled at 7.0 ± 0.5 mg L^{-1}. During the granulation process, the water samples were collected from the reactors at the end of each cycle and the residual concentrations of aniline, COD, and nutrients were determined. The sludge was also sampled and the morphology as well as properties were tested.

2.2. Feeding

Synthetic wastewater was used for development of AGS in R_L and R_H with the ALR of 0.6 and 1.8 kg aniline m^{-3} d^{-1}, respectively. The corresponding OLR was 1.44 and 4.32 kg COD m^{-3} d^{-1} for R_L and R_H, respectively. The compositions of influents were as follows (per liter): aniline 200 mg for R_L and 600 mg for R_H, KH_2PO_4 21.9 mg, $K_2HPO_4 \cdot 3H_2O$ 36.8 mg, $CaCl_2$ 138.75 mg, trace mineral solution 1 mL. The trace element solution was the same as our previous study (Jiang et al., 2016a). The phosphorus in the influent was used for maintaining microbial growth. The pH of the influents was adjusted to 6.5 ± 0.5. The chemical reagents used for the feeding were purchased from the National Medicines Corporation Ltd. of China and were of analytical grade.

2.3. Microbial community analysis

A total of six sludge samples were obtained from the two reactors for analyzing microbial community succession during AGS formation. Sample G_L-1, G_L-2, and G_L-3 were collected from R_L on day 1, 11, and 41. G_H-1, G_H-2, and G_H-3 were harvested from R_H on the same days. The three samples obtained from each reactor represented original sludge, premature sludge aggregates, and mature AGS, respectively. Total genomic DNA was extracted by using the PureLink® Genomic DNA kit (Germany). The integrity, purity and concentration of the DNA were