Chemical Engineering Journal 218 (2013) 253–259

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Journal

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

Aerobic sludge granulation facilitated by activated carbon for partial nitrification treatment of ammonia-rich wastewater

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HIGHLIGHTS

- Sludge granulation was achieved for nitritation treatment of ammonia wastewater.
- Initial GAC addition accelerated the formation of nitritation sludge granules.
- Granulation helped control ammonia oxidation at the partial nitrification level.
- AOB instead of NOB were enriched in nitrifying granular sludge.

ARTICLE INFO

Article history:
Received 7 June 2012
Received in revised form 10 December 2012
Accepted 14 December 2012
Available online 20 December 2012

Keywords:
Aerobic granulation
Partial nitrification
Activated carbon
Activated sludge
Biological nitrogen removal
Wastewater treatment

ABSTRACT

Although the use of partial nitrification, or nitritation, for nitrogen removal via nitrite is an energy-saving method for treating high-strength ammonia wastewater, its stable operation with sufficient enrichment of ammonia-oxidizing bacteria (AOB) is difficult to maintain in activated sludge systems. In this study, an aerobic granulation technique was developed for the effective and stable nitritation treatment of ammonia-rich inorganic influent. Granular activated carbon (GAC) or powdered activated carbon (PAC) was added to the bioreactor to enhance the granulation of slow-growing AOB. The results show that aerobic granules could be formed for partial nitrification through the selective discharge of small and slow-settling sludge flocs, with or without activated carbon addition. However, dosing GAC into the sludge greatly accelerated the granulation process and shortened the granulation period from about 6 weeks to less than 3 weeks with the formation of large and fast-settling granules. In contrast, dosing PAC led to the slower formation of smaller granules. Compared to activated sludge flocs, sludge granulation with selective sludge discharge was found to help halt ammonia oxidation to the level of partial nitrification rather than complete nitrification. Based on the molecular analysis, aerobic granulation resulted in AOB enrichment and the reduction of nitrite-oxidizing bacteria (NOB) in granules, which is highly favorable to a stable partial nitrification operation.

1. Introduction

Nitrogen removal is one of main objectives in wastewater treatment. Partial nitrification for biological nitrogen removal (BNR) via nitrite has recently gained interest because of its considerable energy and cost savings compared to complete nitrification [1,2]. It has become a particularly attractive option for treating high-strength ammonia wastewater with low organic content. The nitrite produced by ammonia-oxidizing bacteria (AOB) in partial nitrification can be readily removed via denitrification by anaerobic ammonium oxidation (anammox) or other similar processes [3,4]. Partial nitrification or nitritation (ammonia oxidation to nitrite) can be achieved in activated sludge systems [5,6] or biofilm reactors [7,8]. However, problems have been reported with the stability of the partial nitrification system because of the accumulation of nitrite-oxidizing bacteria (NOB) in the biomass under the nitrite-rich condition [9,10]. A strict operating condition such as a low DO level (<1.5 mg/L) is commonly requested. In addition, a sufficient washout of NOB is essential to stable nitritation [6,11], which typically requires a short sludge retention time (SRT). However, a longer SRT (>10 d) is commonly required for activated sludge to ensure a high biomass concentration and efficient and reliable treatment performance.

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http://dx.doi.org/10.1016/j.cej.2012.12.044

Aerobic granulation is a process in which loose sludge flocs are transformed into dense granules. Due to attributes such as a compact structure and fast settling velocity, granule formation allows a higher sludge concentration that increases the loading capacity of biological wastewater treatment systems [12]. The selective discharge of loose and slow-settling flocs has been found to be crucial to the transformation from activated sludge to granular sludge [13,14]. Granulation helps maintain and enrich the slow-growing, nitrifying bacteria in a reactor to enhance ammonia oxidation [15]. However, due to the slow growth rates of nitrifying bacteria, complete granulation is rather difficult to achieve in nitrification or partial nitrification, particularly for concentrated ammonia feeds with little organic substrates [16]. In some of the successful cases that have been reported, a long start-up period of 2 months or more was needed to achieve sludge granulation [17–19]. Other factors such as dissolved oxygen, sludge discharge and ammonia concentration can affect the granule formation and performance of ammonia oxidation [20,21]. Nonetheless, effective start-up strategies need to be developed to accelerate granule formation for reliable granulation and stable partial nitrification.

Activated carbon (AC) has a large specific surface area and a fast settling velocity. Dosing with AC enhances aerobic granulation under unfavorable conditions, such as low substrate concentrations and low loading rates [22]. Because AOB have a slow growth rate, it is difficult to increase their concentration in a bioreactor. With the use of AC, AOB would attach to the AC and avoid washout from the reactor during the start-up of granulation. As a result, rapid granule formation could be achieved to ensure the stable operation of nitritation. In this study, laboratory experiments were conducted with three batch bioreactors to cultivate granular sludge for partial nitrification. Granular activated carbon (GAC) or powdered activated carbon (PAC) was added to the sludge mixture in one of the reactors. The aims of this study were to develop an effective technique for the rapid granulation of AOB sludge for nitritation, to investigate the microbial population change during the granulation process and to examine the capability and stability of partial nitrification by the granular sludge.

2. Materials and methods

2.1. Experiment set-up

Three laboratory bioreactors, B1, B2 and B3, were used to grow granular sludge for partial nitrification. Each reactor was a small column (H 30 cm × i.d. 3.6 cm) with a working volume of 200 mL. The reactors were inoculated with nitrifying activated sludge as the seed sludge that had been cultivated in a lab-scale fermentor (Sartorius Biostat®, A Plus, Germany). The feed into the fermentor, named B0, was an NH_4Cl solution with a NH_4^+–N concentration of 200 mg/L and no organic content [23]. The initial sludge concentration added to the column reactors was around 2000 mg/L in terms of mixed-liquor volatile suspended solids (MLVSS).

The bioreactors were dosed with two typical types of AC, GAC and PAC, with the goal of enhancing the sludge granulation process. The GAC and PAC had mean sizes of 224.4 μm and 50.5 μm, respectively, with an apparent density of 1.183 g/cm^3 according to the supplier (Merck, NJ, USA). No AC was added to reactor B1, 0.1 g of GAC was added to B2 and 0.1 g of PAC was added to B3 for an initial GAC or PAC concentration of 0.5 g/L. Aeration was supplied from the bottom of each column by an air pump at a flow rate of 8 L/min to keep the DO concentration in the sludge suspension within a range of 2–4 mg/L. The reactors were fed once every 12 h. The influent to the reactors was a synthetic wastewater prepared with NH_4Cl and KH_2PO_4 without any organic substrates added. Clean seawater collected from Cape d'Aguilar Marine Reserve, Hong Kong, was filtered with a 0.22 μm membrane and added to the synthetic wastewater at a ratio of 1:2 to increase the wastewater salinity. The synthetic wastewater had a salinity of about 1% that is similar to the saline wastewater in Hong Kong. The wastewater influent contained an NH_4^+–N concentration of 400 mg N/L and a PO_4^{3-}–P concentration of 40 mg P/L, which resulted in a volumetric N loading of 0.8 g N/L d in the reactors. The pH of the mixed liquor in the reactors was controlled at around 7.5 during the experimental period by adding a diluted $NaHCO_3$ (0.1 M) solution automatically.

The discharge of small and slow-settling sludge flocs was conducted at the end of each 12-h cycle from each of the column reactors. Before sludge discharge, the sludge was allowed to settle in the column without aeration for a period from 1 to 5 min, depending on the sludge's settling properties and the targeted sludge discharge rate. The slow-settling sludge in the top 40 mL suspension was then removed from the reactors. The sludge concentration in each reactor was measured; accordingly, the amount of daily sludge discharge was adjusted to maintain a biomass MLVSS concentration of around 2000 mg/L in each reactor. After the selective sludge discharge, the remaining sludge suspension was allowed to settle in the column for another 30 min, and the supernatant (~180 mL) was then withdrawn as the effluent from the reactor. The feed solution was added into each reactor to restore the original volume of 200 mL.

2.2. Determination of the nitritation capability of the granules

After the completion of aerobic granulation, the granular sludge was characterized for its partial nitrifying kinetics and settling velocity, in comparison to the seed nitrifying activated sludge. For each sludge sample from B0, B1, B2 and B3, the nitrification and nitritation capability test was performed in a 100-mL glass beaker and the sludge and NH_4^+–N concentrations were 2 g MLVSS/L and 400 mg/L, respectively. The sludge mixture was sampled at various time intervals. The NH_4^+–N, NO_2^-–N and NO_3^-–N concentrations in the liquid phase of the sludge were measured, and subsequently the specific rate of ammonia degradation by the granules was determined.

2.3. Analysis of microbial population and identification of dominant species in the bioreactors

The microbial population of the seed sludge and the mature granules from the three reactors was analyzed. The genomic DNA of the sludge was extracted from the cells using a beadbeater (Mini-beadbeater™, Biospec, Bartlesville, OK, USA) and microcentrifuge (MiniSpin plus, Eppendorf, Hamburg, Germany) [24]. The bacterial 16S rDNA gene sequence (V3 region, corresponding to positions 341–534 of the *Escherichia coli* sequence) was amplified by polymerase chain reaction (PCR) (PTC-200, MJ Research, Waltham, MA, USA) following the previously detailed procedure [25]. The PCR-amplified DNA products were then separated by denaturing gradient gel electrophoresis (DGGE) through 8% polyacrylamide gels with a linear gradient of 30–50% denaturant using the DCode™ Universal Mutation Detection System (Bio-Rad, Hercules, CA, USA). The gels were run for 6 h at 130 V in a 1 × TAE buffer at 60 °C. Afterward, the gels were stained with ethidium bromide for 10 min and then visualized by a UV illuminator. The DGGE images were acquired using the ChemiDoc (Bio-Rad) gel documentation system and the DGGE profile was analyzed by "QuantityOne" (Version 4.6.3, Bio-Rad, Hercules, CA, USA).

A 16S rRNA gene sequence clone library constructed for the seed sludge was used to identify the phylogeny of the DGGE bands of the sludge samples [23]. The sequences of the clones used as markers