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Treatment of municipal sewage with low carbon-to-nitrogen ratio via a novel integrated process

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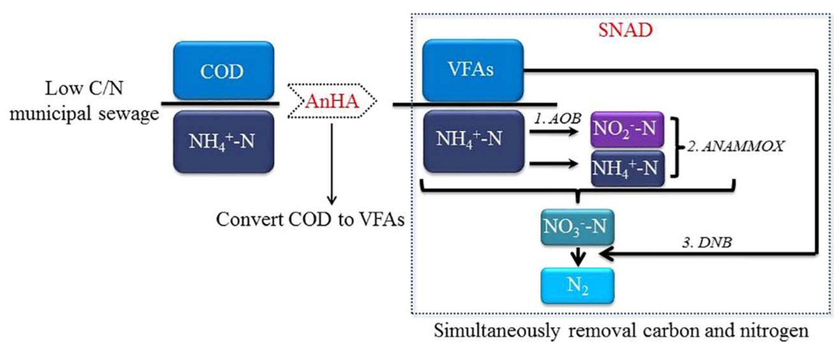
HIGHLIGHTS

- AnHA-SNAD was proposed to treat low C/N municipal sewage.
- The removal rates of COD and NH_4^+-N in AnHA were 27.15% and 1.45%, respectively.
- The removal rates of COD and TN in SNAD were 83.12% and 79.13%, respectively.
- FISH analysis showed the dominant bacterial group of AnHA-SNAD.
- The energy consumption of AnHA-SNAD was accounted for 53.2% of A^2/O.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, a novel integrated process combining simultaneous partial nitrification, anammox, and denitrification (SNAD) with anaerobic hydrolysis and acidification (AnHA) was proposed for the treatment of municipal sewage with low carbon-to-nitrogen ratio (C/N). The average removal efficiencies of chemical oxygen demand (COD) and NH_4^+-N in AnHA were 27.15% and 1.45%, respectively. Meanwhile, the removal rates of COD and total nitrogen in SNAD were approximately 83.12% and 79.13%, respectively. Fluorescence in situ hybridization results showed that acidogenic bacteria accounted for 92.4% of all bacteria in AnHA. The dominant bacterial group in the aerobic layer of the SNAD biofilm was ammonium-oxidizing bacteria (65.13%), whereas that in the anaerobic layer was anammox (47.17%) and denitrifying (38.91%) bacteria. Compared with the energy consumption of the anaerobic–anoxic–oxic process (100%), the AnHA-SNAD energy consumption was only 53.2%, which indicated an energy-saving alternative process for low C/N municipal sewage treatment.

1. Introduction

For municipal wastewater treatment plants (MWWTPs), nitrogen and carbon should be simultaneously removed to meet discharge standards. Ammonium, which is usually the main nitrogen compound in wastewater, is aerobically oxidized to nitrite by ammonium-oxidizing bacteria (AOB) and to nitrate by nitrite-oxidizing bacteria (NOB). These compounds (nitrite and nitrate) can be reduced to nitrogen gas

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using organic matter as carbon sources by denitrifying bacteria (DNB). Therefore, oxygen and organic carbon are required in conventional biological treatment processes. However, municipal sewage usually exhibits a low carbon-to-nitrogen ratio (C/N) [1,2], and the costs associated with external carbon addition and high energy consumption have resulted in the development of alternative processes [3,4].

Simultaneous partial nitrification, anammox, and denitrification (SNAD) process contributes to the possibility of resolving the preceding challenges mentioned [5]. In SNAD, a partial amount of ammonium is converted into nitrite by AOB under low oxygen concentration (Eq. (1)). Subsequently, anammox bacteria convert surplus ammonium and nitrite into nitrogen gas and a small amount of nitrate (Eq. (2)), because anammox bacteria exhibit closer affinity to nitrite than DNB [6]. Then, DNB convert nitrate into nitrogen gas by organic carbon consumption (Eq. (3)). Thus, the coexistence of AOB, anammox bacteria, and DNB in a single reactor has opened possibilities for the simultaneous removal of nitrogen and carbon with low energy consumption and operational costs [7,8]. Owing to the inhibition of organic matter on anammox bacteria [9], pretreatment of municipal sewage is necessary before introducing SNAD [10].

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + H_2O + 2H^+ \quad (1)$$

$$NH_4^+ + 1.32NO_2^- \rightarrow 1.02N_2 + 0.26NO_3^- + 2H_2O \quad (2)$$

$$5CH_3COOH + 8NO_3^- \rightarrow 4N_2 + 6H_2O + 10CO_2 + 8OH^- \quad (3)$$

Generally, the biological removal of 1 mg of NH_4^+-N requires several-fold mg of chemical oxygen demand (COD). COD need to be converted to volatile fatty acids (VFAs) first, and then can be utilized by DNB. So, the effective compounds of carbon sources for DNB are in fact VFAs [11]. Recent studies provide insights into the occurrence possibility of the anammox process in the presence of VFAs, particularly C_2 and C_3 fatty acids. The tolerance level of anammox bacteria for VFAs is higher than that for other carbon sources [12]. So, VFAs are suitable carbon sources for SNAD. As is well known that anaerobic hydrolysis and acidification (AnHA) can convert COD to VFAs, and anaerobic processes have been recognized as a potential energy-saving treatment process for MWWTPs [13]. Thus, a novel integrated process combining SNAD with AnHA, which is used to pretreat municipal sewage to provide feasible carbon sources (i.e., VFAs) for SNAD, is expected to realize sustainable and energy-saving contaminant removal in low C/N municipal sewage.

In this study, AnHA-SNAD was proposed for the treatment of municipal sewage with low C/N to simultaneously remove nitrogen and carbon. First, AnHA and SNAD were set up in biofilm reactors to explore the performance of the integrated process. Second, the microbial community was investigated via fluorescence in situ hybridization (FISH) to understand the biological foundation of AnHA-SNAD. Finally, the energy consumption of AnHA-SNAD was analyzed to discuss the application prospect of the novel integrated process. The successful application of AnHA-SNAD will offer a great future potential for savings of resource and energy in MWWTPs for efficient nitrogen and carbon removal.

2. Materials and methods

2.1. Wastewater

The feeding medium used in this experiment included synthetic wastewater and municipal sewage. The influent synthetic wastewater of AnHA contained sucrose, $(NH_4)_2SO_4$, and KH_2PO_4 as nutrients, whereas the influent synthetic wastewater of SNAD mainly contained COD, NH_4^+-N, NO_2^--N, and PO_4^{3-}-P in the form of CH_3COOH, $(NH_4)_2SO_4$, $NaNO_2$, and KH_2PO_4, respectively. The composition of trace element solutions was based on previous studies [2,14]. The influent pH values were adjusted through the addition of NaOH and H_2SO_4. Municipal sewage was collected from a local MWWTP (Dalian, China), and the characteristics are shown in Table 1. Municipal sewage was pretreated via AnHA to supply a suitable carbon source for SNAD. Then, the effluent of AnHA was discharged to SNAD to simultaneously remove nitrogen and carbon.

Table 1
Main characteristics of municipal sewage.

COD mg/L	BOD_5 mg/L	NH_4^+-N mg/L	TN mg/L	pH	SS mg/L
150–200	60–90	25–30	35–40	7.5–7.8	70–140

2.2. Seed sludge

The inoculation material used in AnHA was obtained from a local MWWTP (Dalian, China), with an initial mixed liquid suspended solid (MLSS) concentration of 6000 mg/L. The seed sludge of SNAD included AOB and anammox bacteria, with an initial MLSS concentration of 4000 mg/L and 5000 mg/L, respectively. AOB was cultivated in a laboratory-scale sequencing batch reactor. The activated anammox bacteria, which account for approximately 89% of Planctomycete-type bacteria and belong to *Candidatus Brocadia anammoxidans*, originated from a laboratory-scale anammox up-flow anaerobic sludge blanket reactor [15].

2.3. Experimental setup

Biofilm reactors used in this study were made of polymethyl methacrylate. As shown in Fig. 1, AnHA was set up in an up-flow non-woven biofilm reactor (UNBR). A total of six non-woven porous polyester paddings (each with an area of 0.01 m^2) were vertically inserted into the reactor to enhance bacteria attachment due to their good adsorption characteristics [16]. The effective volume of the UNBR was 3.2 L. A stirrer was used for mixing, and a subsequent sedimentation tank was equipped with a scraper to prevent accidental sludge loss. The UNBR was hermetic and maintained at room temperature.

As shown in Fig. 2, SNAD was carried out in a non-woven rotating biological contactor (NRBC), which was also filled with the non-woven porous polyester padding as a disc. A total of 15 discs were mounted on a horizontal shaft fixed with stainless steel and driven by an electric motor. The effective volume of the NRBC was 2.5 L. The discs were rotated at 2 rpm to mix the substrate. The liquid inside the reactor was

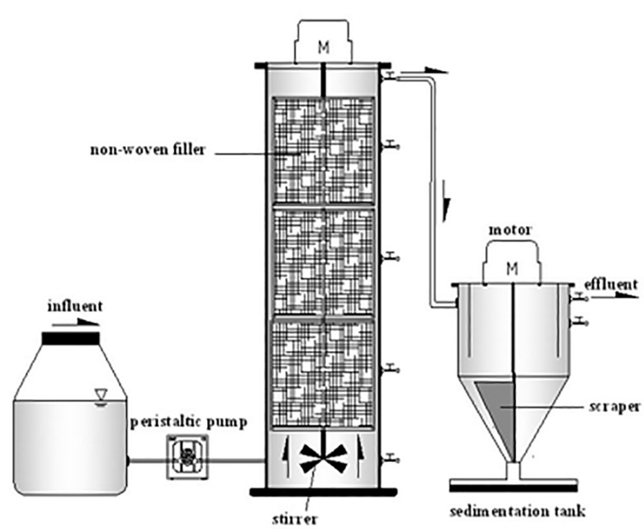

Fig. 1. The sketch of the UNBR for AnHA.