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First full-scale nitritation-anammox plant using gel entrapment technology for ammonia plant effluent

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ABSTRACT

The first full-scale nitritation-anammox process using gel entrapment technology was evaluated for nitrogen removal. Ammonia plant effluents contained not only ammonium but also methanol, an anammox inhibitor. The nitrogen concentration and loading design were 690 mg/L and 400 kg-N/d, respectively. A nitritation reactor (170 m^3) filled with nitrification gel carriers was started up on day 20, with a resulting nitrification efficiency of 58%. A stable nitritation performance was observed without significant nitrate production for >1 year. A nitrogen conversion rate of 3.2 kg-N/m^3/d was observed on the anammox reactor (100 m^3) filled with anammox gel carrier on day 69. The full-scale anammox reactor could be started up in approximately 2 months. Subsequently, stable nitrogen removal performance was observed for >1 year. The average nitrogen loading, nitrogen conversion rate, and total nitrogen removal on the anammox reactor were 3.6 kg-N/m^3/d, 3.0 kg-N/m^3/d and 330 kg-N/d, respectively. The nitrogen removal performance obtained in this study will facilitate further application of the anammox processes to many types of industrial wastewater treatment.

1. Introduction

Biological treatment of nitrogen wastewater is a cost effective method of removing ammonium nitrogen from ammonium-rich wastewater. The conventional denitrification process combining nitrification and heterotrophic denitrification has been widely used globally. However, the conventional denitrification process is difficult to apply to ammonium-rich wastewater with a low organic substrate, such as digester supernatant and ammonia plant effluents. Under these circumstances, additional organic carbon sources (methanol) have to be supplied to facilitate the denitrification process, thereby incurring increased operation cost.

The anoxic ammonium-oxidizing (anammox) reaction is a recently discovered microbial denitrification pathway [1–3], and its application to many kinds of wastewater treatment has recently been reported [4]. During the development of the denitrification process using anammox, immobilization of anammox bacteria is an important requirement because the growth rate and biomass yield of anammox bacteria are very low. Previous studies maintained anammox biomass as granules, following which it was used in the upflow anaerobic sludge blanket (UASB) and sequencing batch reactor (SBR) type reactors [5–7]. A rotation biological contactor has also been applied to immobilize the anammox biomass on the biofilms [8]. In previous applications, the anammox biomass was attached to the surface of the plastic carriers (Kaldnes) and then installed in the moving bed biomass reactor (MBBR) type reactors [9]. We previously developed gel entrapment techniques to immobilize and grow anammox bacteria in an MBBR type reactor [10,11]. Immobilized anammox bacteria carried in a gel are easily separated from the effluent using a screen, and this yields prolonged biomass retention times even with short hydraulic retention times (HRTs). Therefore, a stable and high nitrogen removal performance could be obtained by gel entrapment technology [12,13]. In addition, the seed anammox sludge for the start-up of the anammox reactor could be saved because anammox bacteria could be immobilized inside the gel carriers without the loss of biomass. Considering the aforementioned advantages, the gel entrapment technique was used for the implementation of a full-scale nitrogen wastewater treatment system applied to industrial effluents from an ammonium plant.

During the denitrification process using anammox, half of the ammonium contained in the influent is required to be oxidized to

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Table 1
Characteristics of ammonium plant effluent.

	Designated	Measured				
		AVERAGE	MIN		MAX	
NH_4-N	690	658	583	–	770	mg/L
NO_2-N	0	0	0	–	0	mg/L
NO_3-N	0	0.1	0	–	0.2	mg/L
Total Nitrogen	690	692	612	–	792	mg/L
Methanol	200–400	N.M	N.M	–	N.M	mg/L
TOC	–	37	17	–	49	mg/L
Inorganic Carbon	–	514	455	–	752	mg/L

nitrite using a partial nitritation process [14–16] because nitrite and ammonium are both required for the anammox reaction. Thus, two-stage nitritation–anammox processes were applied for autotrophic denitrification [6,17].

Full-scale anammox plants have been mainly installed for municipal wastewater treatment, such as those of digester supernatant [4]. There are few reports on full-scale anammox plants for industrial wastewater treatment [17]. The present report is the first assessing the performance of nitrogen removal of ammonia plant effluents using the anammox reaction. Ammonia plant effluents have been found to contain not only high concentrations of ammonium but also high concentrations of methanol. Methanol shows considerable toxicity to substrates for anammox bacteria [18]. Therefore, a pretreatment process for methanol removal should be combined with the nitritation-anammox process.

The faster start-up of the nitritation-anammox process using gel entrapment technology and long-term stability of the nitrogen removal performance were clearly demonstrated in the present report.

2. Materials and methods

2.1. Ammonia plant *effluents*

Nitrogen containing wastewater was continuously drained from an ammonium plant at a flow rate of 580 m^3/d. The characteristics of the effluents from an ammonium plant are shown in Table 1. The designated ammonium concentration was 690 mg/L. The methanol concentration was normally around 200 mg/L; however, the methanol concentration was occasionally increased to around 400 mg/L. The average measured ammonium concentration was 658 mg/L, and almost no nitrite and nitrate were detected. The original wastewater did not contained any organic substrates expect methanol. Because the water temperature of the original effluent from the ammonia plant was >60 °C, it was cooled to 30 °C by a heat exchanger using sea water as a coolant before feeding it into the wastewater treatment system.

2.2. Full-scale setup of the nitritation-anammox plant

Fig. 1 shows a schematic diagram of the full-scale plant. This system mainly consists of three parts. First, a denitrification (DN) and biochemical oxygen demand (BOD) oxidation (BD) reactor were used for the treatment of methanol to prevent methanol inhibition of anammox activity. Nitrite for denitrification on the DN reactor was provided following the nitritation (NT) reactor by a recirculation pump. Second, NT and anammox (AX) reactors were installed for the treatment of ammonium nitrogen. Finally, nitrate produced by the anammox reaction was denitrified on the post heterotrophic denitrification reactor by the addition of methanol. The volume of each of reactor was as follows: 40 m^3 for DN, 65 m^3 for BD, 170 m^3 for NT, 100 m^3 for AN, and 30 m^3 for post-denitrification reactor. Gel carriers were installed in each reactor except the post-denitrification reactor. The volume of gel carriers installed in each reactor was as follows: 4 m^3 for DN, 6.5 m^3 for BD, 34 m^3 for NT, and 20 m^3 for AX reactors. A fixed bed carrier (Biofringe, NET, Japan) was installed in the post-denitrification reactor, and the packing ratio was approximately 40% of the reactor volume. In the AX reactor, the gel carriers were agitated using a paddle type mixer at approximately 7 rpm. A paddle type mixer was also installed in the DN reactor, in which the mixing speed was approximately 25 rpm. The mixing speed was set to a level at which gel carriers would not break. Stainless steel wedge wire screens, with a feature slit width of 1.5 mm, were installed at the front of the effluent line of each reactor to separate the gel cubes from the wastewater. In the present system, no sedimentation tanks were installed to maintain activated sludge because each kind of biomass was individually maintained in the gel carriers in each reactor.

The recirculation rate from the NT reactor was basically set at 0.08 Q when the influent methanol concentration was around 200 mg/L. When the influent methanol concentration was increased, the recirculation rate was set at 0.3 Q to enhance methanol consumption (denitrification) in the DN reactor. The recirculation rate was adjusted so that approximately half amount of methanol in the influent was consumed in the DN reactor, and the remaining methanol was fed to the subsequent BD reactor. The pH value in the NT reactor was maintained at 7.5 using NaOH solution, whereas that in the AX reactor was adjusted to 7.6 by H_2SO_4 solution.

The nitrification rate was controlled by adjusting the aeration rate [19]. The nitrification efficiency in the NT reactor was calculated from the values of ammonium concentrations and total nitrogen for the effluent of the NT reactor measured online ($C_{NH4,out}$ and $C_{TN,out}$, respectively), according to the following equation:

$$\eta_{Nit} = 1 - (C_{NH4,OUT}/C_{TN,OUT})$$

The aeration rate was adjusted to meet the appropriate value of η_{Nit}. The total nitrogen concentration was monitored using a fully automated analyzer (TNC-6200, Toray Engineering Co., Ltd., Japan), and the ammonium concentration was monitored using an ammonia electrode (Ammolizer, SCAN, Austria).

2.3. Gel carriers

2.3.1. Denitrification, BOD oxidation, and nitritation gel carriers

The initial gel carriers for denitrification, BOD oxidation, and nitritation used for the full-scale plant were the same gel carriers entrapping activated sludge. Sewage sludge was used as seed sludge because it is easy to obtain and is available in large amounts from sewage treatment plants. Moreover, seed denitrifying bacteria, BOD oxidizers, and nitrifying bacteria were normally included in activated sludge. The suspended solid (SS) concentration of the seed sewage sludge was adjusted at 33 g/L using a centrifuge. Polyethylene glycol (PEG) was used as a prepolymer (Shin-Nakamura Chemical Co. Ltd., Japan). A mixing tank was used to mix the solution of the PEG prepolymer and sewage sludge. Subsequently, a promoter (N,N,N′,N′-tetramethylenediamine) was added into the obtained PEG prepolymer mixture. Potassium persulfate was added to the tank to initiate the polymerization. The resulting polymerized gel was cut into 3-mm cubes. This gel carrier contained 10% (w/v) PEG, 0.5% (w/v) promoter, 0.25% (w/v) initiator, and 2% (w/v) sewage sludge [20]. Fig. 2 shows the photographs of each correct gel carrier from each reactor.

2.3.2. Anammox gel carrier

The enriched anammox sludge was used as seed sludge. The immobilization procedure for anammox bacteria is similar to the aforementioned gel carriers, except that sewage sludge was not