STUDIES ON NITROGEN REMOVAL PERFORMANCES FOR SINGLE STAGE NITROGEN REMOVAL USING ANAMMOX AND PARTIAL NITRITATION (SNAP) PROCESS

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ABSTRACT

The single-stage nitrogen removal using anammox and partial nitritation (SNAP) process was developed as an economical nitrogen removal option for ammonium rich wastewaters. Experimental studies for the evaluation of SNAP treatment performances were conducted using a novel fixed bed reactor employing net-type acrylic-fiber biomass carrier. SNAP process could be successfully applied for the treatment of synthetic wastewater containing high NH_4-N of 500mg/L and maximum T-N removal rate of 0.86kg-N/m^3/d was obtained. Influent TOC of 30 mg/L did not give bad effect on nitrogen removal performances of SNAP process. Influent wastewater containing 10g-Cl^-/L caused 35% of inhibition to nitrogen removal capabilities and the T-N removal efficiencies decreased from 80% to 52% under T-N loading rate of 0.5 kg-N/m^3/d. SNAP treatment capabilities were decreased under operational temperature of 25℃. Reactor DO concentrations ranging from 2 to 3mg/L were proved to be favorable for SNAP treatment.

KEYWAORDS

Anammox, ammonia removal, partial nitritation, single stage nitrogen removal, SNAP

INTRODUCTION

Anammox reaction, which is the recent addition to traditional nitrogen cycle, was confirmed by researchers of Delft University of Technology on 1995(1). Many research works had been focused on the cultivation of extremely slow growing anammox bacteria. Successful establishment of anammox processes were shown to be possible experimentally using sequential batch reactor (SBR)(2), fluidized bed reactor(3) and fixed bed reactor(4). Three actual anammox plants combining partial nitiritation reactor were constructed in Netherland for the treatment of ammonium rich wastewaters.(5) Combining partial nitritation and anammox reactions, ammonium is converted ultimately to gaseous nitrogen by two sequential reactions(6): partial nitritation (Eq. 1) and anammox (Eq. 2).

$$2\ NH_4^+ + 1.5\ O_2 \rightarrow NH_4^+ + NO_2^- + H_2O + 2\ H^+ \quad \text{(Eq.1)}$$

$$1\ NH_4^+ + 1.32\ NO_2^- \rightarrow 1.02\ N_2 + 0.26\ NO_3^- + 2\ H_2O \quad \text{(Eq. 2)}$$

$$1\ NH_4^+ + 0.85\ O_2 \rightarrow 0.44\ N_2 + 0.11\ NO_3^- + 1.43\ H_2O + 1.14\ H^+ \qquad \text{(Eq. 3)}$$

The combination of two conversion steps (Eq. 3) can be done in separate reactors or in a single reactor. Typical systems with separate reactors include Sharon-anammox(7) and partial nitritation-anammox. In order to reduce the footprint of reactors and operational costs for separate process, single-stage nitrogen removal processes such as OLAND(8) and CANON(9), in which anammox and partial nitiritation occurred in single reactor, were developed recently.

During our experiments on separate partial nitritation process using an attach-immobilized reactor packed with a net type acryl resin fiber carrier, an unexpected nitrogen removal was observed. We named this new annmonium removal process as SNAP (Single-stage Nitrogen removal using Anammox and Partial nitritation)(10). In order to elucidate the treatment performances of this SNAP process, treatment capabilities under different operational conditions were evaluated experimentally.

MATERIALS AND METHODS

1) Experimental set-up

The SNAP treatment performances were studied in a reactor system that was designed for control of pH, temperature and aeration rate, as shown in Fig. 1. The reactor was made from acryl resin and had a liquid volume of 4.65 L. A hydrophilic net-type acryl resin fiber material (BX, NET Co., Ltd.; Japan), shown in Fig. 2, was used as the biomass carrier. Some properties of the BX material are shown in Table 1. 10-11 g of BX was stretched on aluminum pipe as shown in Fig. 2 and was set in the reactor.

Table 1 Properties of acryl-fiber biomass carrier

Parameter	Unit	Value
Specific yarn length	m/m^3	23,324
Specific surface area	m^2/m^3	146,5
Yarn diameter	mm	2
Specific weight	kg/m^3	980

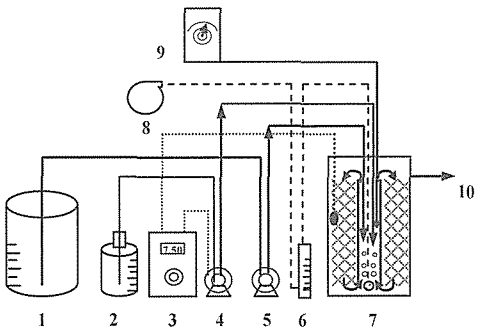

Fig. 1 Schematic diagram of reactor.
(1) Influent tank (2) $NaHCO_3$ solution (3) pH controller (4) $NaHCO_3$ pump (5) Influent pump (6) Airflow meter (7)Reactor (8) Air pump (9) Temp. controller (10)Effluent

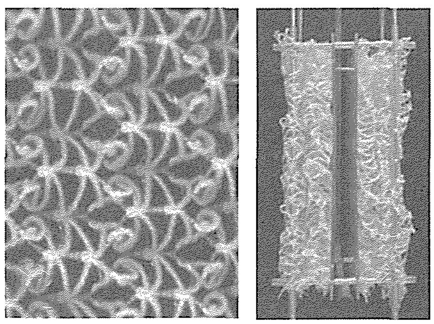

Fig. 2 Photograph of acryl resin biomass carrier.

2) Seed Sludge

The reactor was seeded with 5g (as MLSS) of nitrifying activated sludge. This sludge had been cultivated using a synthetic medium (containing peptone, meat extract, $NaHCO_3$, $MgSO_4$, $CaCl_2$, NaCl, and KCl) by the fill-and-draw method for a long time under total oxidation conditions. The seed sludge was rich in heterotrophic bacteria and nitrifiers. The reactor was fed with an inorganic medium containing NH_4Cl (76.3-1145 mg/L), $NaHCO_3$ (126 mg/L), KH_2PO_4 (44 mg/L), and a micro-mineral solution (1 mL/L) during start-up. The micro-mineral solution consisted of NaCl (1.0 g/L), KCl (1.4 g/L), $CaCl_2.2H_2O$ (1.9 g/L) and $MgSO_4.7H_2O$ (2.0 g/L). During the start-up phase (before day 67), the nitrogen loading rate was increased stepwise by increasing the influent concentration and decreasing HRT.

3) SNAP operation

SNAP reactor was firstly operated to achieve partial nitritation. Suitable operational condition for partial nitritation was determined to be temperature 35℃、pH 7.5-7.7, DO 2-3mg/L under T-N loading rate of 0.5kg-N/m^3/d[11]. Nitrite oxidizing bacteria (NOB) were selectively inhibited under this operational condition. Favorable environmental condition for anammox bacteria enables the proliferation of anammox bacteria inside thick biomass carrier and SNAP process is established. All SNAP experiments were carried out under upper mentioned operational condition.

RESULTS AND DISCUSSION

1) Treatment performances under high influent NH_4-N concentration

SNAP process was discovered during the partial nitritation treatment experiments for influent NH_4-N concentration of 100mg/L[11]. In order to ascertain the treatment capability of SNAP reactor receiving high influent NH_4-N of 500mg/L was evaluated under operational condition of 35℃、pH7.5, HRT 12hours and T-N loading rate of 1.0kg-N/m^3/d. Daily changes in T-N removal rates were shown in Fig. 3. Effluent NO_2-N concentrations were less than 20mg/L during experiment. T-N removal rate was decreased from 83% to 58% on day 25. The reason of this decrease in T-N removal rate was the worse liquid circulation condition caused by the massive growth of SNAP sludge in the reactor. Excess sludge was withdrawn from the reactor on day 35.

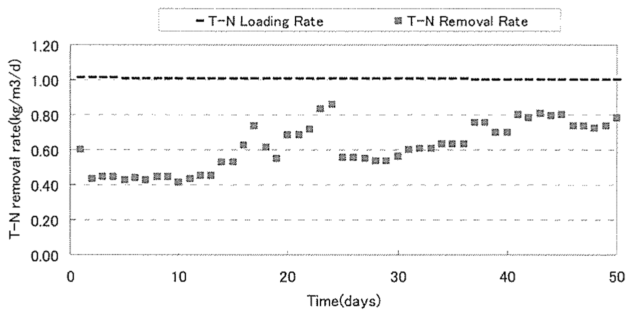

Fig. 3 Daily changes in T-N removal rates

T-N removal rate was recovered to about 80% after day 40. During 50 days of continuous SNAP treatment under high loading rate of 1.0kg-N/m^3/d, average T-N removal efficiencies of 65% was obtained. Maximum T-N removal rate of 0.86kg-N/m^3/d was achieved in this experiment. From this experimental result, our developed SNAP process was proved to be applied for high NH_4-N containing wastewaters such as landfill leachate and anaerobic digester supernatant. This obtained T-N removal rate of 0.86kg-N/m^3/d was proved to be comparable with reported T-N removal rates of another single stage nitrogen removal process of CANON[12] and OLAND[13] processes.

2) Effect of organic matter on SNAP treatment

Both nitrifying and anammox bacteria are autotrophic, so that organic carbon contained in influent wastewater may affect the on SNAP performances. Landfill leachate contains organic carbon except high NH_4-N. Potassium hydrogen phthalate (KHP) and humic acid were used as organic carbon additives to simulate leachate. A concentration ratios of KHP : humic acid = 10:1 was selected to give BOD/COD of about 0.35. Using this synthetic landfill leachate, effect of influent organic carbon on SNAP process was investigated under operational condition of 35℃, pH 7.6-7.8 and aeration rate of 0.06-0.10vvm.

Fig. 4 shows the treatment performances of the SNAP process before and after adding organic carbon to the influent with a TOC/NH_4-N ratio of 0.10-0.15 under T-N loading rate of 0.6 kg-N/m^3/d. This result showed that both ammonium conversion and nitrogen removal efficiencies increased after addition of organic carbon to the influent. The average ammonium conversion and nitrogen removal efficiencies during the last 60 days of operation were 93.7 ± 5.6 % and 84.4 ± 6.9 %, respectively. TOC removal profiles were shown in Fig. 5. The average influent TOC was 30.0 ± 2.5 mg/L and average effluent TOC was 7.3 ± 0.7 mg/L, resulting in an average TOC removal of 75.7%.

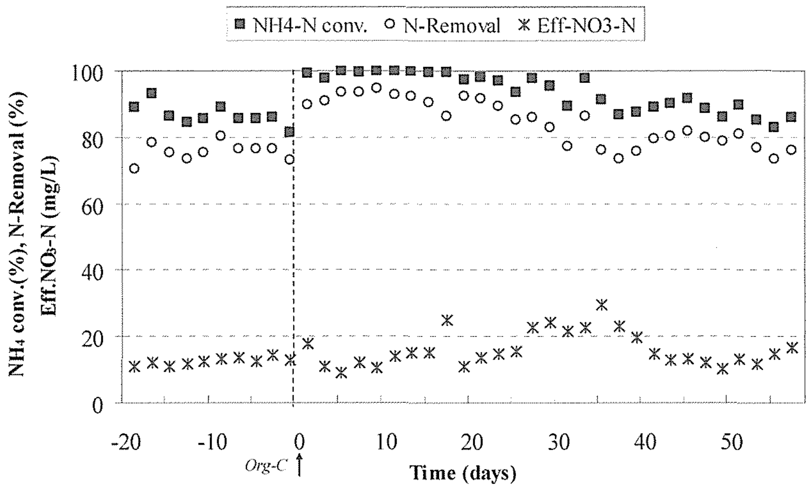

Fig. 4 Treatment performances of SNAP process before and after adding organic carbon to the influent.

An important observation was that effluent nitrate concentrations were almost unchanged after organic carbon addition. This indicates that denitrification did not occur and the anammox reaction was maintained stably. Thus, the increase in nitrogen removal might be attributed to normal heterotrophic denitritation. From this result, high applicability of SNAP process to actual wastewater was demonstrated.

3) Effect of salts concentration

Usually, the target wastewater for SNAP process such as landfill leachate and anaerobic digester liquor contain high chloride components. In general, fresh water bacteria are apt to decrease their activities under high Cl^- concentrations over 10g/L. In order to know the effect of Cl^- on SNAP performances, continuous SNAP treatment experiments using synthetic influent containing high Cl^- components were carried out.

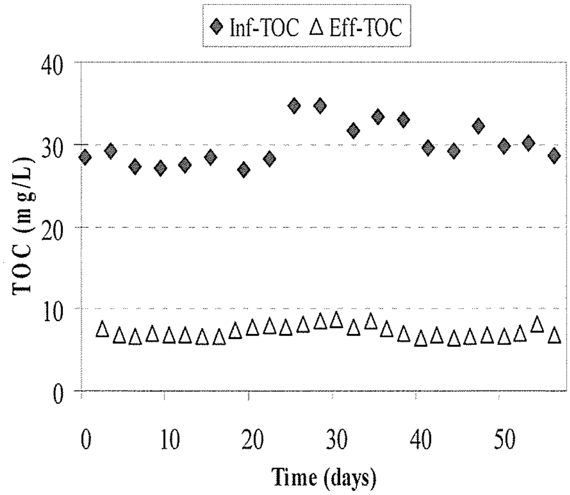

Fig. 5 Daily changes in influent and effluent TOC concentrations during experiments of organic carbon addition

The Cl^- concentration of synthetic wastewater was only 0.72g-Cl^-/L. The continuous SNAP treatment experiments of synthetic wastewater containing 10g-Cl^-/L were conducted. Figs. 6 and 7 showed the daily changes in nitrogen concentrations and T-N removal efficiencies during 90 days of SNAP treatment, respectively.

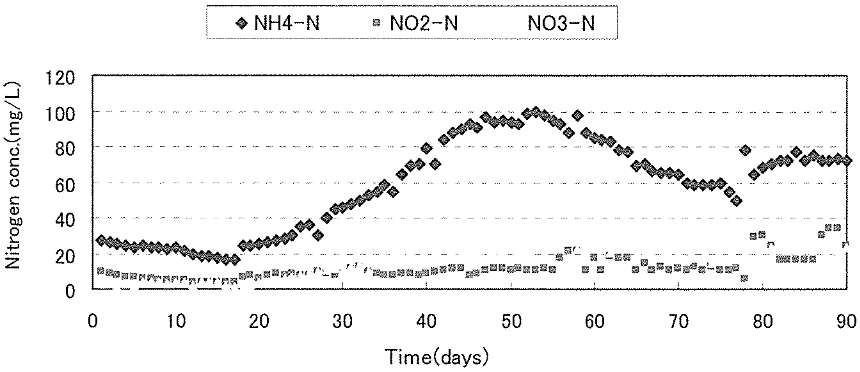

Fig. 6 Daily changes in effluent nitrogen concentrations.

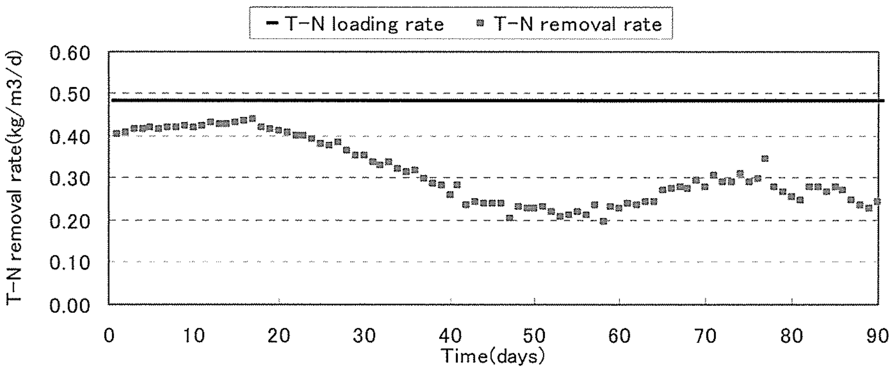

Fig. 7 Daily changes in T-N removal rates under high Cl^- concentrations.

From day 20, Cl^- concentration was increased to 10g/L. Acute inhibition caused by high influent Cl^- concentration was not observed, but T-N removal efficiency of 80% at the beginning experiment decreased gradually and reached to 41% on day 48. Interestingly, effluent NO_2-N concentrations were around 10mg/L during day 20 to 50 over which T-N removal efficiencies were decreasing. On the other hand, effluent NH_4-N concentrations increased gradually from 20mg/L (before increasing Cl^- concentration) to 100mg/L on day 48. This result demonstrates the selective inhibition of ammonium oxidizing bacteria under10g-Cl^-/L. On the contrary, anammox bacteria did not get inhibition by salt concentrations under 10g-Cl^-/L. The decreasing in T-N removal efficiencies stopped on day 50 and the average T-N removal efficiency form day 50 to 90 was 52%. From this experiment, degree of inhibition caused by 10g-Cl^-/L was revealed to be about 35%. Therefore, application of SNAP process to the wastewater containing high salts concentration must be avoided.

3) Effect of temperature

Anammox reaction was proved to show high activities at operational temperature between 20 to 43℃ and its optimum temperature was 40℃(14). Optimum temperature for partial nitritation was reported to 35℃(11). In order to elucidate the effect of operational temperature on SNAP performances, SNAP reactor was operated at 25, 30 and 35℃ under reactor pH of 7.7 and T-N loading rate of 0.4kg-N/m^3/d. Fig. 8 showed the daily changes in T-N removal efficiencies under different operational temperatures. It was clear that T-N removal efficiencies increased with increase in operational temperature. From this result, it was evident that SNAP process should operate at more than 30℃ for obtaining high T-N removal efficiencies. Therefore, temperature control is required for SNAP treatment in the winter season.

4) Effect of DO

Reactor DO affects the distribution of aerobic and anoxic zone inside SNAP biofilm. Effect of reactor DO on SNAP performances was studied by controlling aeration rate. Fig. 9 showed the relationship between aeration rates, effluent nitrogen concentrations and T-N removal efficiencies. Under low aeration rate of 0.01vvm, about 50% of influent NH_4-N was detected in the effluent owing to the insufficient supply of DO to ammonium oxidizing bacteria (AOB). Effluent NH_4-N concentrations decreased with increase in aeration rates, but

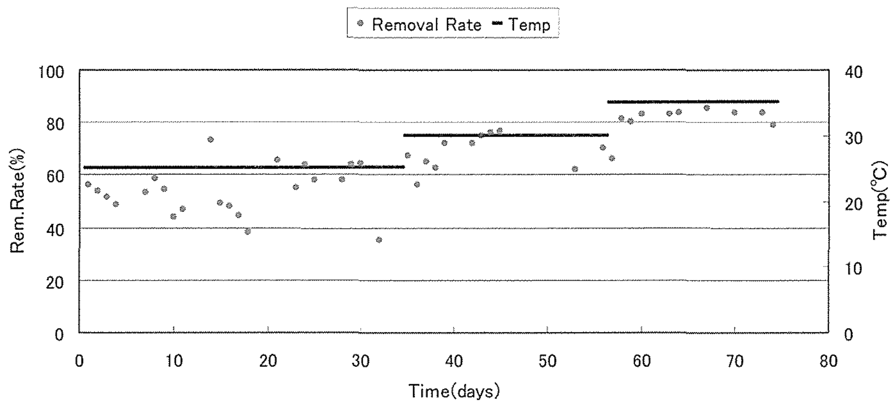

Fig. 8 T-N removal under different operational temperatures.

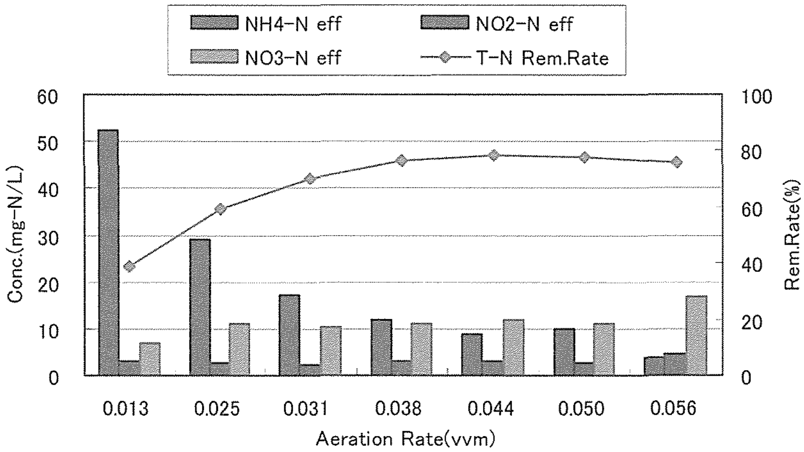

Fig.9 Effect of aeration rate on effluent nitrogen concentrations and T-N removal efficiencies
(T-N loading rate:0.4kg-N/m^3/d, pH:7.7, Temp. 35℃)

nitrification proceeded under aeration rate of 0.056vvm and high NO_3-N concentrations were detected in the effluent. Stable SNAP performances were obtained under aeration rates between 0.014 to 0.05vvm. Reactor DO concentrations ranging from 2.0 to 3.0mg/L were recorded under aeration rates between 0.012 to 0.056vvm.

CONCLUSIONS

Treatment performances of SNAP process under different operational conditions were evaluated experimentally and the following results were obtained.

1) SNAP process was proved to be applied for the treatment of synthetic wastewater containing high NH_4-N concentration of 500mg/L. Maximum T-N removal rate of 0.86kg-N/m^3/d, which is comparable to that for another single stage nitrogen removal processes, was obtained.
2) Through the continuous treatment of synthetic landfill leachate containing KHP and humic acid as organic carbon, it was revealed that influent TOC of 30 mg/L did not give the bad effect on nitrogen removal performances of SNAP process.
3) Influent Cl^- concentration of 10g/L gave 35% of inhibition to nitrogen removal of SNAP process, and the T-N removal efficiency was decreased from 80% to 52%. This decrease in TN removal performances was supposed to be caused by the inhibition of AOB by Cl^-.
4) SNAP treatment capability was decreased under operational temperature of 25℃.
5) Reactor DO concentrations ranging from 2 to 3mg/L were proved to be favorable for proper SNAP treatment.

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