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学 位 論 文 題 目　Fundamental Study on Hydroxyapatite (HAP)-Enhanced One-Stage Partial Nitritation and Anammox (PNA) Process (一槽式 HAP-PNA プロセスに関する基礎的研究)
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論文内容要約

The anaerobic ammonium oxidation (anammox) process for removing nitrogen from wastewater is a focus of research worldwide because of its high energy efficiency and low costs compared with the conventional nitrification and denitrification process. The one-stage partial nitritation/anammox (PNA) process, where aerobic nitritation and anaerobic anammox reactions occur simultaneously in a single reactor, has attracted the attention of many researchers because of its more compact structures and lower investment costs than the two-stage process.

However, many wastewater treatment plants are hesitated and flinching in the face of the application of the anammox process due to the obstacles including unstable operation, low microorganism concentration, fragile in complicated conditions, and so on. Recently, hydroxyapatite (HAP) was found to be a standout granular support medium that produces stronger and more compact granular sludge with greater settleability. This can increase the microorganism concentration in the system, which would require a smaller reactor volume, despite the removal of phosphorus via the precipitation of HAP.

The integrated HAP and one-stage PNA process has been attempted in our laboratory and future application potentials appeared. However, more fundamental studies about the mechanism, key factors and features of the HAP formation are necessary for the wide application of this novel process. Therefore, this study includes the following chapters:

In Chapter 1, the background of the nitrogen and phosphorus pollution and treatment was reviewed. Then, the objectives of this study were proposed. Finally, the contents and constructure of this thesis were introduced.

In Chapter 2, the upgrading of the anammox-based nitrogen removal processes: performance, stability, and control strategies based on the microorganism retaining, process composition and operational performane was reviewed. The HAP-enhanced granular sludge in a one-stage PNA process is promising for high anammox bacteria (AnAOB) content, as well as high nitrogen removal rate (NRR) and nitrogen removal efficiency (NRE).

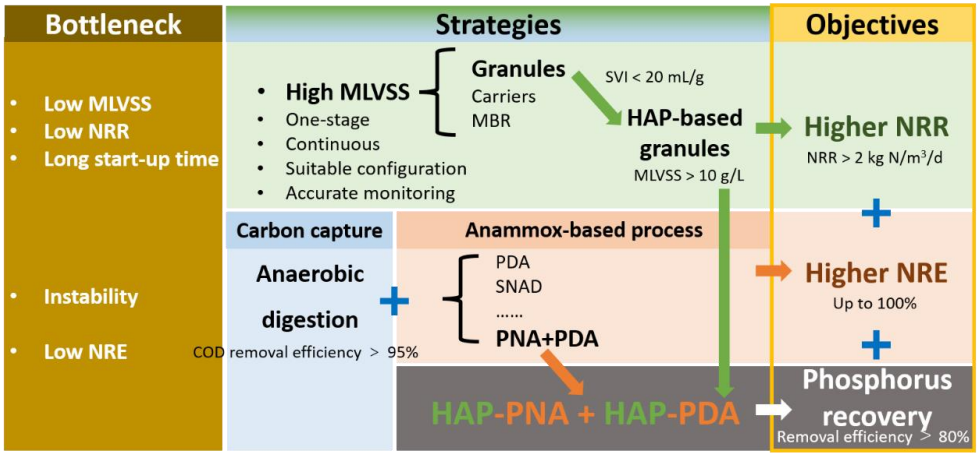

Fig. 1. The bottlenecks, strategies and objectives in the recent anammox process.

In Chapter 3, key factors improving the stability and the loading capacity of nitrogen removal in a HAP-enhanced one-stage PNA process were explored. A stable and high NRR of 2.0 ± 0.1 kg N/m³/d and NRE of 81 ± 3% was achieved, respectively, in a one-stage air-lifting PNA process with HAP-based micro-granules. A phosphorus removal efficiency (PRE) of 77 ± 6% was also obtained when the influent calcium concentration was 120 mg/L. It was found that calcium addition to ensure sufficient HAP formation and a proportionable phosphorus removal rate (PRR)/NRR ratio, 0.02, was necessary to obtain the desirable sludge settleability and a sufficient biomass in the reactor with an increasing NLR. The compact morphological properties of HAP-enhanced sludge and the existence of HAP was also confirmed. HAP-based micro-granules were obtained and a two-layer spatial distribution of functional bacteria was confirmed with FISH. Finally, the observed shift in the AnAOB species was attributed to either the death of some of the original species.

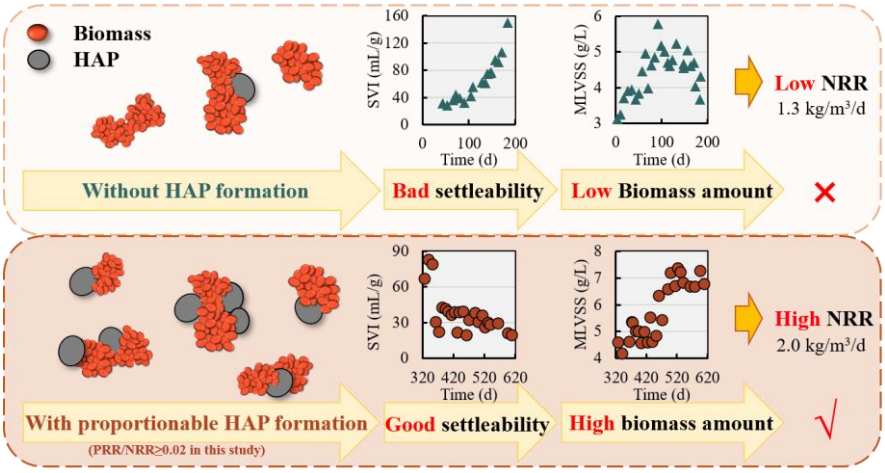

Fig. 2. Efficient improvement of biomass amount and NRR.

In Chapter 4, the nitrogen removal capacity and phosphorus removal performance of the one-stage PNA system with enhancement strategies at a short hydraulic retention time (HRT) were explored. Although the potential of the HAP granular sludge in the improvement of the sludge settlement was shown in las t chapter, how high the nitrogen removal rate can achieve and how an excellent operational performance can be achieved is not known yet. According to the results in this study, the reliability of the process was guaranteed and the nitrogen removal performance was enhanced using the HAP-based granular sludge. A high microorganism concentration of 15 g/L and a high NRR of 4.8 ± 0.5 kg N/m³/d were achieved stably at 25 °C in a one-stage PNA system by forming HAP-enhanced granular sludge. The properties and excellent settleability of the granular sludge were confirmed. The mechanisms of the HAP-based enhancement and requirements (HAP formation, high AnAOB activity, low HRT, selective sludge separation, simultaneous reaction and settlement) for achieving a high NRR were discussed. It is anticipated that the results of this study will provide technical guidance for future research on wastewater management and the applications of more efficient one-stage PNA processes.

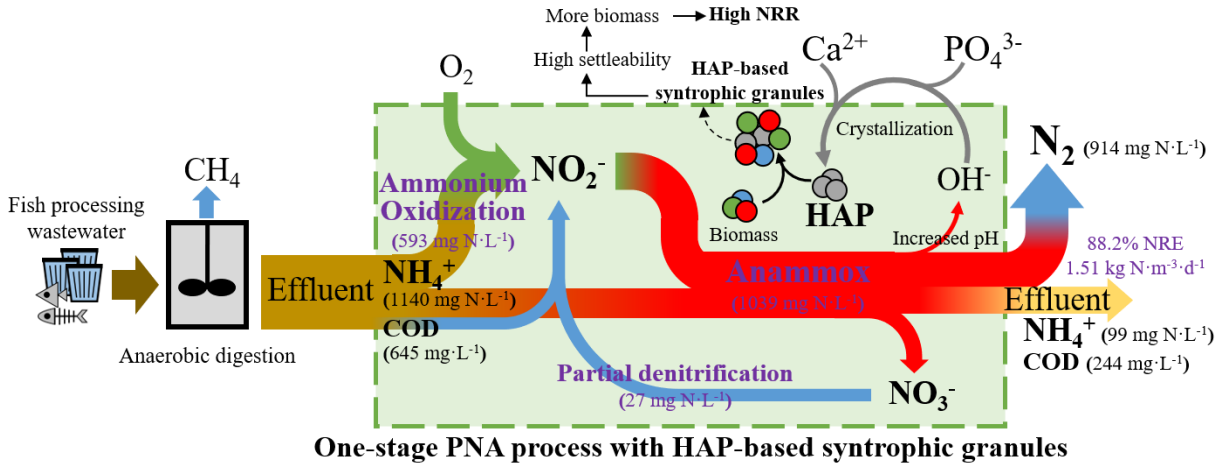

Fig. 3. Mechanism of nitrogen removal from fish processing wastewater by one-stage PNA process.

In Chapter 5, high nitrogen removal performance of anaerobically treated fish processing wastewater by one-stage PNA process with HAP-based syntrophic granules and granule structure was explored. The wastewater with a high ammonium concentration forms due to the high protein concentration in the fish processing wastewater after the anaerobic digestion for the organic matter removal and methane production. Therefore, the nitrogen removal is necessary from the anaerobic-treated wastewater. The nitrogen removal from actual fish processing wastewater was accomplished in a one-stage partial nitritation/anammox process with HAP-based syntrophic granules. A high NRR of 1.51 ± 0.13 kg N/m³/d and a high NRE of 88.2%

were achieved. Anammox bacteria of *Ca.* Kuenenia stuttgartiensis and AOB of *Nitrosomonas* were the predominant bacteria. HAP was detected by analyzing the compounds and the distribution of inorganic elements in the sludge. Finally, the reaction kinetics in the granules was determined. This study shows the high efficiency and potential of this process for the treatment of the fish processing effluents from an anaerobic reactor.

In Chapter 6, nitrogen removal by a HAP-enhanced micro-granule type one-stage PNA process following anaerobic membrane bioreactor treating municipal wastewater was studied. The treatment of municipal wastewater with low nitrogen concentration via PNA process is still difficult. However, in order to achieve the sustainable society with low greenhouse gas emissions, low energy consumption and low waste production, it is attracting to use anammox-based process treating the municipal wastewater. Nitrogen was removed successfully from a pilot AnMBR treated municipal wastewater by a HAP-enhanced micro-granule type one-stage PNA process. An NRE of 80%, an NRR of 0.36 kg $N/m^3/d$ and a COD removal efficiency of 54% were achieved stably in this novel PNA process. The NOB was effectively inhibited and washed out via the combined free ammonia shock strategy with low dissolved oxygen and the HAP-enhanced granules. The HAP-enhanced granules guaranteed the stable nitrogen removal performance by maintaining a high biomass at a low HRT. Overall, the combination of AnMBR and this PNA process provided a promising engineering solution for sustainable municipal wastewater treatment.

In Chapter 7, general conclusions and perspectives in the future were summarized. The system including anaerobic digestion, HAP-PNA and HAP-partial denitratation/anammox was proposed for more efficient nitrogen removal and phosphorus recovery. However, the relationship between AnAOB and denitrifiers should be appropriately adjusted via the influent C/N ratio to ensure the stability of anammox-based systems. Moreover, the requirements of HAP formation include suitable pH and enough calcium concentration in the influent. The required HAP formation rate or calcium addition should be decided based on the influent nitrogen concentration, the phosphorus concentration and the HRT.

These studies focused on the mechanism and application of HAP-enhanced one-stage PNA process. The excellent nitrogen removal performance was obtained by using the HAP-enhanced granular sludge in the one-stage PNA process, providing a more promising choice and the enhancement strategies for the improvement of the sludge and operational performance in the future application of anammox-based process.