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In-situ restoring nitrogen removal for the combined partial nitritation-anammox process deteriorated by nitrate build-up

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ABSTRACT

The nitrogen removal performance in the combined partial nitritation-anammox (CPNA) process was seriously deteriorated by the nitrate build-up. The purpose of this study was to develop and optimize an in-situ restoring strategy based on hydroxylamine (NH_2OH) dosing and solids retention time (SRT) control for the deteriorated CPNA process. Results showed that the 0.55 kgN m^{-3} d^{-1} of nitrogen removal rate could be recovered by 20 mgNH$_2$OH L^{-1} of hydroxylamine dosing and 40 days of SRT control, the nitrate concentration in effluent was decreased from the highest 548.4 mgN L^{-1} during deterioration to 65.1 mgN L^{-1} after restoration, and the ratio of NO_3^--N$_{produced}$/NH_4^+-N$_{consumed}$ in one SBR cycle was reduced from the highest 87.0% to 9.13% finally. The inhibition of nitrite-oxidizing bacteria (NOB) by NH_2OH dosing alone was reversible because the nitrate build-up occurred again from 106.9 to 287.6 mgN L^{-1} within just 11 days after NH_2OH dosing was stopped. The evolution of the anammox bacteria, ammonium-oxidizing bacteria (AOB) and NOB from quantitative PCR (qPCR) assays verified the changes of the nitrogen removal performance of the CPNA process and proved that this in-situ restoration strategy could successfully solve the problem of nitrate build-up in the CPNA process.

1. Introduction

The anaerobic ammonium oxidization (anammox) is a novel, low-cost alternative to the conventional nitrogen removal process [1,2], especially for treating wastewater with high-strength ammonium and low C/N ratio produced from many industries, e.g., semiconductors, pharmacies, and anaerobic digestion of substrates which are rich in protein [3–6]. Among many configurations of the full-scale processes based on anammox, the installations of one-stage combined partial nitriation-anammox (CPNA) process have become more popular [7–9] due to the less investment and operation costs, simpler reactor control and operation, better prevention of nitrite inhibition and advantage on reducing N_2O emission [10,11]. However, it was reported that numerous full-scale CPNA systems faced the problem of nitrate build-up in the effluent reaching to more than 100 mgN L^{-1} [12], which was caused by the bloom of nitrite oxidizing bacteria (NOB). For example, at Zürich-Werdhölzli WWTP, Switzerland, after 2 years of successful operation, the reactor was found to have developed quite a strong NOB population, as evidenced by the effluent nitrate concentrations increasing to over 200 mgN L^{-1} (>30% of the influent ammonium) [12]. At Plettenberg WWTP, Germany, increasing nitrate concentrations were observed in the effluent of a deammonification plant after stable operation for several months, finally the overall nitrogen removal rate was decreased from 80% to 40%, and the growing activity of NOB was proved to be the immediate cause [13]. Therefore the key of controlling nitrate build-up for successful application of the CPNA process is the effective suppression or wash out of NOB, because high population of NOB in the suspended biomass or biofilm system will break down the stable partial nitritation resulting in the nitrate build-up, and then deteriorate nitrogen removal performance [8,14,15]. Besides, the growth of anammox bacteria will be limited seriously as high population of NOB competes for nitrite with the anammox bacteria.

To deal with the nitrate build-up problem in CPNA process caused by the NOB, several strategies have been reported in the previous researches. Re-inoculation with the anammox biomass devoid of NOB was the primary solution for not only the problem of NOB but also the inhibition of anammox bacteria [5,9,12]. Though large amount of the anammox seeding sludge is easy to obtain in Europe since most of the full-scale anammox projects worldwide have been in operation there [7], however, it still needs

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large investment for other places in the world to start the anammox engineering applications from scratch where the demand of new nitrogen removal technologies is urgent, as the cultivating period of the anammox bacteria is extremely long or the costs of importing the anammox seeding sludge from abroad directly is very high. On the other hand, the increasing nitrate concentrations might be observed again in the effluent after re-inoculation if the system operated with the same behavior as before [13]. Hence, re-inoculation is not a universal solution to the problem of nitrate build-up. The second approach was to wash out the NOB from system by controlling a 1.5 days of solids retention time (SRT) as in the Sharon process [16]. But this method is not applicable for the CPNA process with the suspended biomass system because the anammox bacteria have a slower growth rate than the NOB's [17]. Given the specific SRT, the eliminating rate of anammox bacteria would be much higher than that of the NOB. In addition, the CPNA process with the biofilm system is also an exception since the biofilm can sustain microorganisms with very different growth kinetics [18], thus it is difficult to selectively wash out the NOB from the CPNA process. Another approach for dealing with the problem of NOB was to reduce the oxygen supply which favored the growth of ammonium oxidizing bacteria (AOB) [19], but results showed that there was no significant selective advantage for AOB in the CPNA process when the dissolved oxygen (DO) concentrations decreased to as low as 0.2 mg L^{-1} [13]. Though intermittent aeration could restore the partial nitritation based on the concept that the NOB needed a longer lag phase after transition from anoxic to aerobic phases to fully ramp up their metabolism under aerobic conditions [20], however, it still took long period (6 months) to successfully suppress the NOB growth [13]. Though high nitrate concentration occurred in the CPNA process can be in-situ removed through denitrification, this approach needs more organic carbon sources (mainly methanol) which will not only affect the anammox bacteria growth but also offset the cost-saving advantage of the anammox. Results from Güven et al. [21] clearly showed that methanol was the most potent inhibitor for the anammox bacteria, and could lead to the complete and irreversible loss of anammox bacteria activity with its concentrations of as low as 0.5 mM. Recently, a cyclone device was introduced to the DEMON process for enrichment of anammox bacteria [22]. The suspended sludge flocs could be removed from the overflow of the cyclone, so the NOB in flocs were removed from the system effectively and the enrichment of the anammox biomass could be realized by recycling the downflow of the cyclone device back to the DEMON process. However, when facing the problems of high NOB population and low activity of anammox bacteria coupling with poor granulation in the CPNA process simultaneously, whether or not the cyclone still can selectively wash out the NOB and enrich the anammox bacteria is a question with no positive answer till now. To the best of our knowledge, there are no effective solutions to overcome the problem of nitrate build-up caused by high NOB population once and for all in the deteriorated CPNA process at present, and the need of a feasible strategy to in-situ restore the impaired nitrogen removal performance is urgent.

Hydroxylamine (NH_2OH) is an intermediate in both nitrification and anammox processes, and shows particular effects on the nitrifying and anammox bacteria, respectively [23–25]. For example, addition of hydroxylamine at low concentrations in the autotrophic nitrifying biofilm system could inhibit the growth of NOB irreversibly, meanwhile the partial nitritation of ammonium to nitrite was also promoted [26,27]. In aerobic nitrifying granule system with DO > 5 mg L^{-1}, stable partial nitritation was successfully achieved at 10 mgN L^{-1} of NH_2OH dosage [28]. Researches conducted with the anammox biomass also showed that NH_2OH addition could stimulate the removals of ammonium and nitrite simultaneously, and recover the anammox activity [17,29,30]. Therefore, the authors supposed whether or not it was possible to develop an in-situ restoring strategy based on the NH_2OH for the deteriorated CPNA process with high NOB population. However, this possibility still needs to be examined because no relative researches have been reported the synchronous effects of NH_2OH dosing in the deteriorated CPNA process where the NOB coexists with AOB and anammox bacteria. In addition, it is necessary to confirm whether or not the inhibition of NOB by NH_2OH dosing is still irreversible because both the AOB and anammox bacteria in the biomass can incorporate the NH_2OH into their metabolism processes. This proposed restoring strategy is quite different from the previous researches that the NH_2OH was only dosed separately to the nitrifying biomass or the anammox system. If the inhibition of NOB by NH_2OH dosing is reversible, is it effective to control the NOB bloom through a combination of dosing NH_2OH and wasting sludge and finally solve the nitrate build-up problem in the CPNA process?

Therefore, the purpose of this study was to explore and optimize an NH_2OH dosing strategy for in-situ restoring the nitrogen

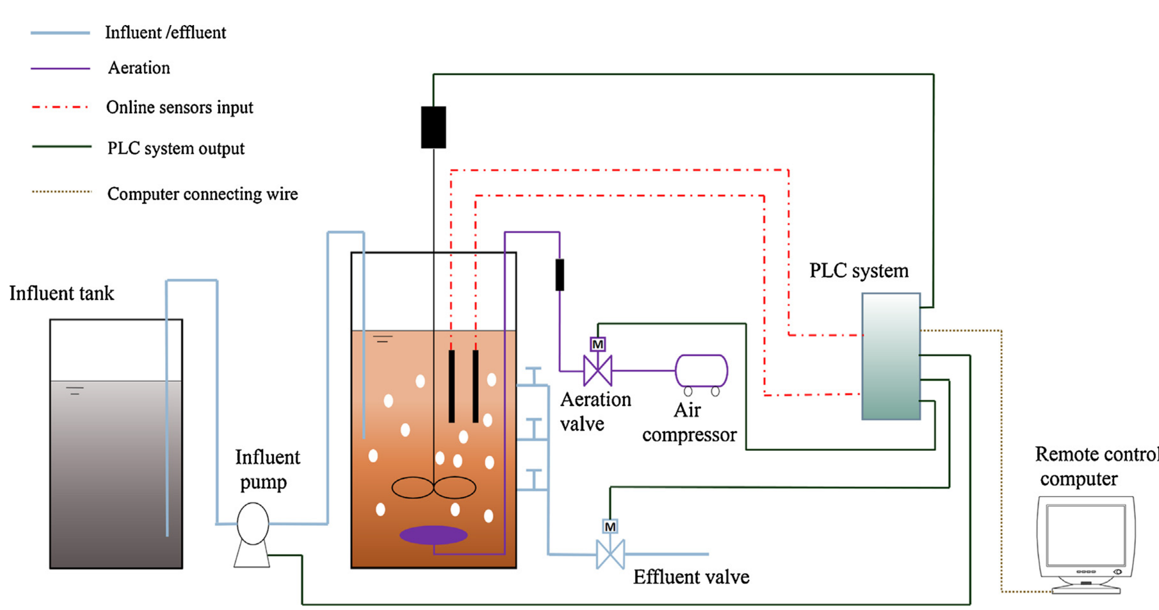

Fig. 1. Schematic diagram of the CPNA process.