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MODELLING, SIMULATION AND OPTIMIZATION OF AUTOTROPHIC NITROGEN REMOVAL PROCESSES

MODELLEREN, SIMULEREN EN OPTIMALISEREN VAN AUTOTROFE STIKSTOFVERWIJDERINGSPROCESSEN

door

ir. Stijn Van Hulle

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Rector: **Prof. Dr. A. De Leenheer**

Decaan: **Prof. Dr. ir. H. VAN LANGENHOVE**

Promotor: **Prof. Dr. ir. P. VANROLLEGHEM**

Summary

For the treatment of ammonia-rich streams, such as reject water of an anaerobic sludge digester, the combination of the ANaerobic AMMonium OXidation (Anammox) process with a process for partial nitrification such as the SHARON or OLAND process can offer a more sustainable alternative to a conventional nitrification-denitrification process. Mathematical models can help to optimize this so-called autotrophic nitrogen removal process. The goal of this research was therefore to develop such a model and with this aim in mind the research was divided into six parts.

After a short introduction part, the second part (chapter 2) summarizes the state-of-the-art knowledge concerning autotrophic nitrogen removal into a literature review. The influence of different factors such as temperature, pH and dissolved oxygen on the activity of nitrifying and Anammox organisms was discussed and an overview is presented of the different reactor configurations that have already been evaluated.

In a third part (chapters 3.1 and 3.2) a model was constructed for the autotrophic nitrogen removal process and implemented in the modelling and simulation environment WEST® (Hemmis NV, Kortrijk). The model was based on the ASM1 model and has as key feature that the uncharged components ammonia (NH_3) and nitrous acid (HNO_2) and not the charged components ammonium (NH_4^+) and nitrite (NO_2^-) are considered as the actual substrate and inhibitor of ammonium and nitrite oxidizers. This approach was confirmed in the experimental study in this thesis (chapter 4.2).

The fourth, experimental, part was started with chapter 4.1. In this chapter the start-up and operation of a lab-scale SHARON reactor in view of its coupling with an Anammox reactor is discussed. Also, several practical experiences with the SHARON reactor were presented. One concerns the evaporation of water from a lab-scale reactor. This evaporation can amount to 20% of the influent flow rate and in this way leads to a higher sludge age than expected from standard chemostat analysis. As inoculum for the SHARON reactor sludge from an SBR system treating synthetic domestic wastewater was used. This sludge was not adapted to the conditions prevailing in the SHARON reactor. A fast start-up procedure directly imposing the “normal” operating conditions of the SHARON reactor, such as an influent total ammonium concentration of 1000 mgTAN-N L^{-1}, a temperature of 35°C and a hydraulic and sludge residence time of 1 day, resulted in the wash-out of the ammonium oxidizers and thus failure of the process. A slow start-up procedure (in semi-SBR mode) allowing the nitrifying sludge to slowly adapt to the conditions prevailing in the SHARON reactor turned out to be the best method to start-up. Stable operation was achieved after 30 days.

After the successful start-up of the reactor, the influence of different TAN influent concentrations and different influent total inorganic carbon:total ammonium nitrogen (TIC:TAN) ratio's was tested. As theoretically predicted a higher TIC:TAN ratio in the influent resulted in a higher TNO_2:TAN ratio in the effluent of the SHARON reactor. This TNO_2:TAN ratio is very important as the Anammox process requires an almost 1:1 TNO_2:TAN ratio for proper operation.

In chapter 4.2 the assumption that ammonia (NH_3) is the actual substrate and nitrous acid (HNO_2) is the actual inhibitor for ammonium oxidation was confirmed by batch tests with sludge originating from the SHARON reactor. With these batch experiments it was furthermore possible to determine the kinetic parameters of ammonium oxidation, such as the ammonia affinity constant, as well as the direct influence of temperature and pH on the growth rate of ammonium oxidizers.

The experimental work was concluded with chapter 4.3, in which a robust titrimetric method for simultaneous determination of TNO_2 and TAN was developed. For nitrogen concentrations typical for the SHARON reactor (350-750 mgN L^{-1}) results from this titrimetric method and a colorimetric method (Dr Lange GmbH, Germany) were in close agreement and had similar measurement accuracies. For typical nitrogen concentrations in the Anammox reactor (0-100 mgN L^{-1}) only the TAN concentrations agreed. The TNO_2 concentration could not be measured with the desired accuracy, as the concentrations were too low.

The fifth part focussed on model applications and started with chapter 5.1 in which the start-up and operational data of an OLAND membrane bioreactor were compared with model simulations. A good agreement was obtained between measured and simulated TAN, TNO_2, nitrate, O_2 and particulate COD concentrations. With the calibrated model the influence of temperature, hydraulic and sludge residence time on the operation of the reactor was examined. The simulation study showed that under oxygen-limited conditions an Anammox-suited effluent could be obtained by fine-tuning the aeration, even at lower temperatures (e.g. 20°C).

In chapter 5.2 the model-based interpretation of start-up and operation of an Anammox reactor is discussed. For this study sludge from a full-scale domestic wastewater treatment plant was used as inoculum. Quantitative data, i.e. TAN, TNO_2 and nitrate concentrations were in good agreement with model simulations. The simulations predicted a gradual increase of Anammox organisms and this was confirmed by qualitative data. The sludge colour changed over the experimental period from brownish to reddish, a typical colour of Anammox biomass. Also an increasingly positive signal with an Anammox specific FISH probe (AMX820) was observed. Further FISH analysis showed that the Anammox organisms were of the type "Kuenenia stuttgartiensis".

In chapter 5.3 an estimate was made of the time necessary for start-up of an Anammox reactor as function of the influent concentration, temperature, hydraulic and sludge residence time, bearing in mind that no Anammox inhibition was considered.

The limited knowledge of the kinetic parameters of the Anammox process is seen as one of the most important bottlenecks for further model simulation studies. Therefore in chapter 5.4 a basis for the further experimental determination of the parameters was laid out based on sensitivity analysis and optimal experimental design. From this study it became clear that among the nitrogen species only the measurement of TNO_2 and TAN yields information on the parameters, while no information can be gained by measuring nitrate and nitrogen gas. Further, at normal operation of the reactor no information can be obtained concerning the TNO_2 inhibition constant and hence a dedicated experiment to gain more information was proposed. This experiment consisted of injecting a TNO_2 solution such that the mixed liquor concentration reaches 30 mgTNO_2-N L^{-1}.

In chapter 5.5 the interaction between Anammox and competing processes such as heterotrophic growth is studied in a biofilm reactor. The most important factors determining the Anammox activity in a biofilm are the oxygen transfer to the biofilm, the influent TAN and COD concentration, TNO_2 inhibition and influent dynamics. Application of autotrophic nitrogen removal in a single biofilm reactor was found to be limited by the amount of oxygen that can be transferred to the biofilm. Systems with two reactors (partial nitrification reactor and Anammox reactor) suffer less from this transfer limitation and therefore seem the better option for treating more concentrated streams. From the simulations it also became clear that the presence of COD is beneficial for the removal of TAN due to the occurrence of simultaneous denitrification. Dynamic influent conditions also lead to lower Anammox activity because of inhibition by TNO_2 and oxygen. In a 2-reactor system the first (partial nitrification) reactor can act as a buffer for the Anammox reactor. Hence, the effect of dynamic conditions will be less detrimental in a 2-reactor configuration.

In a sixth and final part the main conclusions of the work were generally discussed and perspectives for further work were formulated. A first interesting topic for future research is the further investigation of the Anammox growth and decay kinetics. Especially the magnitude and form of the TNO_2 inhibition should be determined. Also, it should be investigated which concept, decay or maintenance, is most appropriate to model decay of Anammox organisms. A second main topic for future research is the use of the mathematical model describing autotrophic nitrogen removal for simulation and control of full-scale systems and evaluate what the benefits of dedicated sludge reject water treatment are for the overall plant.