

51 (2013) 4228–4234 May



Model-based evaluations of operating parameters on CANON process in a membrane-aerated biofilm reactor

Leila Vafajoo*, Mahdi Pazoki

Islamic Azad University, South Tehran Branch, Chemical and Environmental Engineering Group, Deh-haqi St., Ahang Blvd., Tehran, Iran Tel. +98 2133010400; email: vafajoo@azad.ac.ir

Received 7 February 2012; Accepted 2 July 2012

ABSTRACT

Nitrogen compounds, such as ammonium, naturally appear in most wastewaters necessitating treatment in order to prevent oxygen depletion and eutrophication of surface water bodies. Conventional biological nitrogen removal from wastewater usually performed using possible set-ups of sequential aerobic nitrification and anoxic denitrification processes. The completely autotrophic nitrogen removal over nitrite (CANON) process is a combination of partial nitrification an Anaerobic Ammonia Oxidation (ANAMMOX) in which the aerobic ammonium oxidizers and ANAMMOX bacteria perform two sequential reactions under oxygen-limited conditions. CANON process is suitable to remove ammonium from wastewaters characterized by the low content of organic carbon yet abundance of ammonium. It performs with limited volume and reasonable budget; hence, any available nitrification unit might be converted into a new improved one rather easily while benefiting from flexibility of CANON control strategies. To better understand such process' behaviour, one might initially start with developing a mathematical model as a useful tool. In this research, the modelling of CANON process was demonstrated in a Membrane Aerated Biofilm Reactor (MABR) through the ASM3 reference model. It was shown that, with 0.7 mm biofilm thickness the optimal nitrogen removal might be obtained, when the ammonium concentration in influents reached 130 gN/m^3 and DO equaled to $1.3 \text{ gO}_2/\text{m}^3$.

Keywords: CANON; Membrane-aerated biofilm reactor (MABR); ANAMMOX; ASM3 pattern; Mathematical modelling

1. Introduction

For many years in the past, N_2 cycle microbiology was mainly used to improve agricultural yields. About 40 years ago, the potential of this cycle in removing nutrients from wastewater was discovered and studies were aimed towards this direction. Nitrogen appears in wastewater in several forms including (i) Free and ionized ammonia (NH_3 and NH_4^+), nitrate (NO_3^-) and nitrite (NO_2^-) as well as; (ii) organic compounds. Although nitrogen is an essential nutrient for biological growth and is one of the main constituents in all living organism, an excessive presence of it in the effluent wastewater should be avoided. In nitrification process, nitrite is generated as an intermediate compound.

*Corresponding author.

The Sixth Dubrovnik Conference on Sustainable Development of Energy, Water and Environment Systems (SDEWES 2011), 25–29 September 2011, Dubrovnik, Croatia

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The operating microorganisms in this reaction are Nitrobacter (NB) and Nitrosomonas (NS) types and perform the reaction in two phases [1,2], that is,

$$2NH_4^+ + 3O_2 \xrightarrow{NS} 4H^+ + 2NO_2^- + 2H_2O$$
(1)

$$2NO_2^- + O_2 \xrightarrow{NB} 2NO_3^-$$
(2)

Nitrification of wastewater is usually engineered to be followed by denitrification. Denitrification is a biological reduction process for nitrate removal from a nitrified effluent, since de-nitrifying bacteria are heterotrophic, an oxidizable carbon source is essential for this process. Methanol has been widely used for this purpose [2]. The Anaerobic Ammonia Oxidation (ANAMMOX) process puts in operation autotrophic conversion of ammonia and nitrite into nitrogen gas on the basis of the following equation [3]:

$$NH_4^+ + 1.3NO_2^- \rightarrow 1.02N_2 + 0.26NO_3^- + 2H_2O$$
 (3)

The bacteria shown to be responsible for the ANAMMOX process belong to the order of planctomycetales. Although the bacteria are anaerobic, their activity is only reversibly inhibited by oxygen. Furthermore, the ANAMMOX bacteria are inhibited by high nitrite concentrations. These bacteria have been enriched from inocula from different wastewater treatment plants and are characterized by a low maximum growth rate and thus have to be grown in a reactor with sufficient biomass retention time. Moreover, these ANAMMOX bacteria have also been detected in several wastewater treatment systems with high nitrogen losses and low input of organic materials [3].

In combined application of partial nitrification and ANAMMOX, less oxygen for nitrification will be needed, since only a part of ammonium is oxidized to nitrite. In addition, adding of an exogenous organic carbon source for such process is not necessary [3] due to the ANAMMOX being performed by autotrophic bacteria [3]. It is noteworthy that the combined process of partial nitrification and ANAMMOX might be performed in a system consisting of either a single reactor or two reactors.

2. CANON process

CANON process was first developed by combining partial nitrification and ANAMMOX in a benchscale experimental process. As both performed reactions in the reactor were autotrophic, CANON process was phrased completely autotrophic nitrogen removal over nitrite. The performed reaction in CANON process is as follows [4]:

$$\begin{split} NH_3 + 0.85O_2 &\rightarrow 0.11NO_3^- + 0.44N_2 + 0.14H^+ \\ &+ 1.43H_2O \end{split} \tag{4}$$

Some advantages of single-reactor CANON process in comparison with older methods such as nitrification and denitrification including cost reduction, smaller space needed, less sludge produced, prevention of producing unwanted N₂O and NO products and no needs to add organic carbon to the wastewater. In the biofilm-based CANON process, nitrite oxidizers compete with ammonium oxidizers for oxygen and with ANAMMOX organisms for nitrite in biofilm [5]. Typically, in the CANON process, the NS like ammonia oxidizing bacteria are active in the outer aerobic region of both biofilm and aggregates, while ANAMMOX bacteria are active in the inner anoxic region. In this way, the ANAMMOX bacteria are protected from oxygen which is consumed in the outer aerobic region because oxygen is inhibiting the ANAMMOX activity [5]. The oxygen mass transfer efficiency from gas to the liquid phase and effective biomass retention are considered the two key rate limiting factors for the operation of a CANON system [5]. Further investigations indeed are needed to promote the oxygen mass transfer in a high biomass retention reactor configuration such as the immobilization process. This is an important challenge in order to scale up the CANON system from the laboratory to an industrial application.

Membrane-aerated biofilm reactors (MABR) represent a new technology for aerobic wastewater treatment, in which hydrophobic gas permeable membranes are used for bubble less oxygen transfer [5]. Perhaps, the most important feature of an MABR is its biofilm that is grown on the membrane surface and has fundamentally different stratification compared to natural biofilms. In a membrane-aerated biofilm (MAB), oxygen partially penetrates the biofilm generating an oxygen concentration gradient; hence, the region nearest to the membrane is oxygen rich, whereas the region neighbouring the biofilm–liquid boundary is rather oxygen poor.

Some of the factors affecting the performance of a CANON process in an MABR include the following: biofilm thickness, dissolved oxygen (DO) concentration level, and ammonium surface loading. It is note-worthy that the affinity constants and biofilm density have no effect on such performance [5,6].

3. CANON process mathematical modelling

In this research, the CANON process modelling in a membrane reactor was accomplished on the basis of

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[able]

ASM3 reference model (ASM3 Pattern) as well as Hao et al. [6] and Volcke et al. [7].

The purpose of this modelling was to forecast the ammonium, nitrite and nitrate concentrations, and nitrogen removal efficiency in a membrane reactor towards optimization of the available nitrification and denitrification units of the municipal wastewater refineries. In this venue, nitrogen was removed from the wastewater through the CANON process in an MAB reactor. The list of the CANON process variables considered in the present model represented in the Table 1.

Some modelling hypotheses for the present work are as follows:

- (1) In order to avoid many complicated equations arising due to the temperature effects, the maximum specific growth of ANAMMOX's organisms was assumed constant; hence, its changes with temperature were ignored. Moreover, due to the ANAMMOX's organisms slow and small growth rate, effects of changes of this assumption upon modelling results might well be insignificant. To support this hypothesis, it was previously shown that a temperature of 30 °C affected the ANAMMOX's process activity at a maximum change of specific growth rate of $\pm 0.03 \, day^{-1}$ which was rather realistic as the activation energy of the ANAMMOX's organisms for this was found to be 70 kJ/mol [6].
- (2) Ammonium and nitrate do not inhibit nonetheless; nitrite inhibits the ANAMMOX process. In reality, however, nitrite concentration rarely reaches such a high level to do so. The nitrite inhibition on the ANAMMOX's process was therefore not included in the current model's rate equation [8].
- (3) Since through the available references, the required kinetic parameters for CANON

Table 1		
List of variables	of the CANON process	

Symbol	Description	Unit
S_{O_2}	Oxygen concentration	gO_2/m^3
S _{NH₄}	Ammonium concentration	gN/m^3
$S_{\rm NO_2}$	Nitrite concentration	gN/m^3
S_{NO_3}	Nitrate concentration	gN/m ³
S_{N_2}	Nitrogen gas concentration	gN/m ³
X _{NH}	Aerobic ammonium oxidizers	gCOD/m ³
X _{NO}	Aerobic nitrite oxidizers	gCOD/m ³
X _{AN}	ANAMMOX organism	gCOD/m ³

Stoichiometric matrix for aerob	vic ammoniu	m oxidation, nitr	rite oxidation a	and ANAMMOX					
Process $S_{{ m O}_2}$ (g	O ₂ /m ³) S _{NI}	H ₄ (G N/m ³)	SNO ₂ (g N/m ³)	S _{NO3} (G N/m ³)	S_{N_2} (g N/m ³)	X _{NH} (g COD/m ³)	X _{NO} (g COD/m ³	X _{AN}) (g COD/m	$_{3}^{X_{1}}$ (g COD/m ³)
Aerobic NH ⁺ ₄ oxidation growth of $X_{\rm NH}$ 1 – 3.4	13/Y _{NH} -1/	$M_{\rm NH} - i_{\rm NBM}$	$1/\gamma_{ m NH}$			1			
Aerobic end .resp. $X_{\rm NH}$ –(1 –)	f_{X_1}) i_{NB}	$\mathbf{W} = i\mathbf{W}\mathbf{X}\mathbf{I} \times f\mathbf{X}\mathbf{I}$				-1			fxı
Anoxic end. resp.X _{NH} Aerobic NO ₂ ⁻ oxidation	ⁱ NB	$IX_{j} \times IXN_{j} - M_{i}$		$-(1 - f_{XI})/2.86$	$(1 - f_{XI})/2.86$	-1			fxı
growth of X_{No} 1 – 1.1 Aerobic end resp. X_{No} –(1 –)	$4/Y_{NO} -i_{N}$	JBM = i M V + i V V + i V V + i V	$^{-1/Y}$ NO	$^{1/Y}$ NO			1		fvi
Anoxic end resp. X _{No} ANAMMOX		$IX_{f} \times IXN_{i} - W$		$-(1 - f_{XI})/2.86$	$(1 - f_{XI})/2.86$		-1		fxr
growth of X _{AN}	-1/	$\gamma_{\rm AN} - i_{\rm NBM}$	$-1/Y_{AN} - 0.8$	$3 \ 1/1.14$	$2/Y_{\rm AN}$			1	
Aerobic end resp. $X_{AN} - (1 - j)$ Anoxic end resp. X_{AN}	f _{X1}) ⁱ NB ⁱ NB	$\frac{IXf \times IXNi - Wi}{IXf \times IXNi - Wi}$		$-(1 - f_{XI})/2.86$	$(1 - f_{XI})/2.86$			- 1 - 1	fxı fxı

modelling were experimentally determined only at a temperature of 30 °C and under pH levels of 7 or 8, changes of temperature and pH ignored in order to simplify the resulting equations. Doing so nonetheless is expected to give rise to some discrepancies between this model predictions and experimental data available in the literature.

- (4) Model affinity constants have almost no influence on the total effluent concentration. Two relative affinity factors $K_{O_2}^{NO}/K_{O_2}^{NH}$ and $K_{NO_2}^{NO}/K_{NO_2}^{AN}$ shown to influence the outcome of the competition between ammonium oxidizers, nitrite oxidizers and ANAMMOX organisms.
- (5) In reality, lower rations of the two aforementioned factors of item 4, above, are not very likely to occur; therefore, the present limited knowledge on these affinity constants seems sufficient rendering them reliable to be

utilized in simulations for macroscopic reactor behaviour.

(6) Regarding the tuning of the process parameters, an optimal DO concentration level at which the maximum nitrogen removal occurred was associated with a certain ammonium surface load. Hence, carefully regulating the DO concentration level in the bulk of the liquid became crucial to achieve the optimal process performance.

Ultimately, providing the rationalizations used behind the main developed steps of the current model, the stoichiometric matrix on the basis of ASM3 reference model (ASM3 Pattern) was applied for aerobic ammonium and nitrite oxidation and ANAMMOX according to relationships displayed in Table 2.

Considering this matrix calculation and its relations to reactions velocity, nine coupled Ordinary Differential Equations (ODEs) developed as follows:

$$\begin{aligned} \frac{d_{SO_{2}}}{dt} &= -21.867 \times \mu_{NM}^{MAX} \times \frac{SO_{2}}{K_{O_{2}}^{N} + SO_{2}} \times \frac{S_{NH_{1}}}{K_{NH_{1}}^{NH_{1}} + S_{NH_{1}}} \times X_{NH} - 0.9 \times b_{NH} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{1}} + SO_{2}} \times X_{NH} \\ -26.805 \times \mu_{NM}^{MAX} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{2}} + SO_{2}} \times \frac{S_{NO_{2}}}{K_{NO_{1}}^{NH_{1}} + S_{NH_{2}}} \times X_{NO} - 0.9 \times b_{NO} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{1}} + SO_{2}} \times X_{NH} \\ -26.805 \times \mu_{NM}^{MX} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{1}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NH_{1}}^{NH_{1}} + S_{NH_{1}}} \times X_{NH} - 0.09 \times b_{NO} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{1}} + SO_{2}} \times X_{NH} \\ -26.805 \times \mu_{NM}^{MX} \times \frac{SO_{2}}{K_{O_{1}}^{NH_{1}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NH_{1}}^{NH_{1}} + S_{NH_{1}}} \times X_{NH} + 0.068 \times b_{NH} \times \frac{SO_{2}}{K_{O_{2}}^{NH_{1}} + SO_{2}} \times X_{NH} \\ + 0.068 \times b_{NH} \times \eta \times \frac{K_{O_{1}}^{NH} + SO_{2}}{K_{O_{1}}^{NH_{1}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NO_{1}}^{NH_{1}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NO_{1}}^{NH_{1}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{2}}^{NH_{2}} + SO_{2}} \times \frac{S_{NO_{1}}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{2}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{2}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{2}}^{NH_{2}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{1}} + SO_{N}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{1}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{1}} + SO_{2}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{1}} + SO_{N}} \times \frac{SO_{2}}{K_{NO_{1}}^{NH_{1}} + SO$$

$$\begin{aligned} \frac{dx_{NH}}{dt} &= \mu_{NH}^{Max} \times \frac{SO_2}{K_{O_2}^{NH} + SO_2} \times \frac{S_{NH_4}}{K_{NH_4}^{NH} + S_{NH_4}} \times X_{NH} - b_{NH} \times \frac{SO_2}{K_{O_2}^{NH} + SO_2} \times X_{NH} \\ &- b_{NH} \times \eta \times \frac{K_{O_2}^{NH}}{K_{O_2}^{NH} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{NH} \\ \hline \frac{dx_{NO}}{dt} &= \mu_{NO}^{Max} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_2}^{NO} + S_{NO_2}} \times X_{NO} - b_{NO} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times X_{NO} \\ \hline - b_{NO} \times \eta \times \frac{K_{O_2}^{NO}}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_2} + S_{NO_3}} \times X_{NO} \\ \hline \frac{dx_{AN}}{dt} &= \mu_{AN}^{Max} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{NO} \\ \hline - b_{NO} \times \eta \times \frac{K_{O_2}^{NO}}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{NO} \\ \hline \frac{dx_{AN}}{dt} &= \mu_{AN}^{Max} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{NO} \\ \hline - b_{AN} \times \eta \times \frac{K_{O_2}^{NN}}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{AN} \\ \hline - b_{AN} \times \eta \times \frac{K_{O_2}^{NN}}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{AN} \\ \hline - b_{AN} \times \eta \times \frac{K_{O_2}^{NN}}{K_{O_2}^{N} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{AN} \\ \hline + 0.1 \times b_{NO} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times X_{NH} + 0.1 \times b_{NO} \times \eta \times \frac{K_{O_2}^{NH}}{K_{O_2}^{NH} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{NO} \\ + 0.1 \times b_{AN} \times \frac{SO_2}{K_{O_2}^{NO} + SO_2} \times X_{AN} + 0.1 \times b_{AN} \times \eta \times \frac{K_{O_2}^{NH}}{K_{O_2}^{NO} + SO_2} \times \frac{S_{NO_3}}{K_{NO_3} + S_{NO_3}} \times X_{AN} \\ \end{array}$$

Through solving these equations simultaneously by MATLAB software, the changes in concentrations

Table 3 Wastewater composition and characteristics utilized in this study

Wastewater characteristics	Value
NH ₃	$140 \mathrm{gN/m^3}$
NO ₂	$2 \mathrm{gN/m^3}$
NO ₃	60 gN/m^3
Phosphate material	2 gN/m^3
Aerobic ammonium oxidizers	400 g COD/m^3
Aerobic nitrite oxidizer	800 g COD/m^3
ANAMMOX organisms	$600 \text{ g COD}/\text{m}^3$

Table 4

Characteristics of the reactor and operating conditions in this study

Description	Quantity
Bulk volume	$13.2 \mathrm{m}^3$
Biofilm surface	$3,240 \mathrm{m}^2$
Influent flow rate	50 m ³ /day
Ammonium surface load	$2.16 \mathrm{gN/m^2} \mathrm{day}$
Density biofilm	$50,000 \text{ gCOD/m}^3$
Porosity biofilm	$0.75 \mathrm{m}^3$ liquid/m ³ biofilm
Thickness biofilm	0.7 mm
Time of process	120 day

of all elements were obtained. The specifications of the utilized wastewater and reactor in this research were provided in Tables 3 and 4, respectively.

4. Results and discussion

It is well known that, when a system is investigated through development of a mathematical model, such model's coverage for different ranges of input values of different parameters is indeed a priority. Therefore, a model providing more accurate forecast of the system's behaviour due to inputs of different concentrations with lowest errors is considered to be the most desirable one. The provided model in this research was applicable to influent wastewater with



Fig. 1. Comparison of the NH₄ concentration between simulation and experimental results.



Fig. 2. Comparison of the NO_2 concentration between simulation and experimental results.



Fig. 3. Comparison of the NO_3 concentration between simulation and experimental results.

ammonium concentrations ranging from 20 to 200 gN/m^3 , nitrate concentrations ranging from 40 to 80 gN/m^3 and nitrite concentration less than 2 gN/m^3 . Through the following figures, the available experimental and laboratory results were compared with modelling outcomes and shown that the developed model had satisfying ability to predict different concentrations with maximum of 8% mean errors.

Fig. 1 displayed variations of the NH₄ concentration during four months of operations. As it is shown, after a while ammonium concentration will decrease because of its consumptions through both partial nitrification Eq. (1) and ANAMMOX Eq. (3). This diversity at the end of this period will be slower as more ammonium consumed by the partial nitrification compared with its consumed amount for ANAMMOX leading to lowering of the ammonium concentration.

In Fig. 2, the variations of the NO_2 concentration during the four months of operations were provided. As indicated in this figure, at the beginning, the NO_2 concentration raised due to this species being produced by the partial nitrification Eq. (1) process. Then, its concentration was reduced because of consuming the nitrite that was produced by the ANAMMOX process Eq. (3).

In Fig. 3, the variations of the NO_3 concentration during the same four months provided. As observed, the concentration of nitrate will decrease from the very beginning to the end of the process. As the CANON process is accomplished under oxygen-limited conditions, the produced nitrite via this process (i.e. due to the partial nitrification) will not be converted into nitrate because of the shortage of oxygen; therefore, the nitrate concentration will decrease gradually.

It is noteworthy that, the 8% deviation between experimental and theoretical data, although being rather satisfactory, was most probably due to assuming some parameters such as temperature and pH levels to be constant or some other model simplifications implemented in the velocity equations. Moreover, this 8% deviation was an average value determined for all results generated from the present model. In other words, it is expected that by utilizing optimum ammonium amounts and considering temperature variations, the aforementioned deviations be further reduced.

5. Conclusions

In this research, modelling of partial nitrification and ANAMMOX process were combined and performed in a reactor under constant volume and operation conditions such as temperature and pH levels. The novelty of this work was developing a mathematical model for such a process. Indeed, such a theoretical case study had not been undertaken previously. The modelling results satisfactorily described the process stoichiometric equations on one hand and the experimental results provided in the literature on the other. The 8% average deviations between these data were due to some simplifications implemented while developing the model in order to avoid highly complicated equations as well as to prevent lack of access to some experimental data. It is a foregone conclusion that this model might be considered a satisfactory first step for forecasting and studying the concentration of all influential parameters involved for such a process during the performance period undertaken.

Acknowledgements

These researchers do thankfully acknowledge the authors of Refs. [5] and [9] whose data were used to confirm the mathematical model developed in this study. 4234

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