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ORIGINAL ARTICLE

Study of performance of nitrification process

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KEYWORDS

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Abstract The objective of this paper was to study the static behavior and the performance of an experimentally validated model for nitrification process using a single reactor high activity ammonia removal over nitrite (SHARON) process. The model consists of mass balances of total ammonium, total nitrite, ammonium oxidizers, and nitrite oxidizers. The steady state analysis allowed the construction of practical diagrams that show the effect of operating conditions (dilution rate and ammonium feed concentration) as well as kinetic parameters on the performance of the bioreactor. The focus is made on the region that allows for the conversion of ammonium to nitrite and the prevention of further oxidation of nitrite to nitrate. The findings of this study are applicable which can delineate the effect of process variables on the performance of the bioreactor.

1. Introduction

Nitrification can be considered to be the single most important process in the development of today's theoretical understanding of biological wastewater treatment processes (Stuven and Bock, 2001). Not only is it an important process in wastewater treatment plants but it served as the basis for the development and modeling of a number of wastewater treatment processes such as activated sludge and biofilm reactors (Nelson and Sidhu, 2009; Sheintich, 1993; Sheintich et al., 1995).

The global biological nitrogen fixation is estimated to be around 250 Mt (Stuven and Bock, 2001). It is estimated that only 10–15% of the fertilizer ends up in food chain. The rest is discarded into the air, soil and ground water, causing serious environmental problems (Stuven and Bock, 2001). Moreover, once nitrogen has been consumed as human food or animal feed, only a small fraction is incorporated into the body, while the major part of the nitrogen is released again into the environment in the form of domestic wastewater. Over the last decades, there are more stringent requirements concerning nutrient discharge levels. Commonly used international regulations impose a level of 10 mg N total/L for effluents from waste water treatment plants. Although some physico-chemical techniques are available for the treatment of nitrogen such as (magnesium–ammonium phosphate, MAP) precipitation or ammonia stripping (Gujer, 2010), these techniques have given the way for the much cheaper and more effective biological treatment techniques.

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The traditional biological method for the removal of nitrogen from wastewaters is carried out using nitrification/denitrification process. In the nitrification process, ammonium, in its dominant form (NH_4^+), is oxidized with oxygen via nitrite (NO_2^-) to obtain nitrate (NO_3^-) using some autotrophic bacteria. During the subsequent denitrification, nitrate is reduced via nitrite to nitrogen gas (N_2) by some heterotrophic bacteria.

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + 2H^+ + H_2O \text{ (ammonium oxidizers)}$$
$$NO_2^- + 0.5O_2 \rightarrow NO_3^- \text{ (nitrite oxidizers)}$$
$$NH_4^+ \text{ (ammonium)} + 2O_2 \rightarrow NO_3^- \text{ (nitrate)} + H_2O + 2H^+$$

Contrary to autotrophic bacteria, that uses CO_2 for carbon source, heterotrophic bacteria needed in the denitrification process requires an organic carbon source such methanol, which should be supplied externally when necessary:

$$NO_3^- \text{ (nitrate)} + 5/6CH_3OH + H^+ \rightarrow 1/2N_2 + 5/6CO_2 + 13/6H_2O$$

The traditional process of nitrification/denitrification over nitrate takes place in activated sludge reactors in the main wastewater treatment plant or in separate units for reject water treatment. Different reactor types are used such as continuous stirred tank reactors (CSTR), sequencing batch reactor (SBR), biofilm airlift suspension (BAS) reactors and membrane bioreactors (MBR) (Forrez et al., 2009; Fux et al., 2006; Manser et al., 2005a,b).

The conventional nitrification/denitrification over nitrate can be applied to ammonium-rich wastewaters, provided that sufficient oxygen is supplied to the system and that sufficient carbon source is available for denitrification. However, the oxidation of high ammonium concentrations causes a significant pH-decrease, that limits further ammonium conversion due to a limitation of the free ammonia (NH_3), being the actual substrate, and due to nitrous acid (HNO_2) inhibition.

Several novel nitrogen biological treatment processes have been developed lately (Forrez et al., 2009; Gujer, 2006, 2010; Salem et al., 2003). The drivers behind such developments are the search for effectiveness and the reduction in aeration costs. One class of these techniques is the nitritation process (Volcke, 2006). The nitritation process is based on the conversion of ammonium to nitrite, while further oxidation of nitrite to nitrate is prevented, thus realizing aeration cost savings, in comparison with conventional nitrification to nitrate. Moreover, less waste sludge is produced. When subsequent denitrification is applied, less carbon source must be added, while sludge and CO_2 productions are decreased.

One subclass of these novel methods is the so called SHARON (single reactor high activity ammonia removal over nitrite) process. In a SHARON reactor (Volcke, 2006) ammonium is converted to nitrite while further conversion of nitrite to nitrate is prevented. This is achieved by operating at high operating temperatures (30–40 °C) and neutral pH and suitable dilution rate. Under these conditions, nitrite oxidizers grow slower than ammonium oxidizers, so they are washed out by setting an appropriate sludge retention time (typically 1 day), preventing nitrate formation. Because of the short retention time, organisms with high activity are selected, that usually also possess a low affinity for ammonium. As a result, the SHARON process is very well suited for reducing the nitrogen load of streams with high ammonium content. Volcke and coworkers in a series of studies (Sbarciog et al., 2006; Volcke, 2006; Volcke et al., 2006, 2007), examined the behavior of the SHARON model for constant pH. The authors examined the existence, uniqueness and stability of the equilibrium points of a SHARON reactor model. It was shown that up to three physical equilibrium points can occur, although in all cases only one equilibrium point is globally asymptotically stable. The effect of changing parameters and input values on the number of equilibrium points and their stability has also been examined. It was shown (Forrez et al., 2009; Pollice et al., 2002) that the performance of the removal process depends on a number of parameters such as levels of dissolved oxygen, pH and organic load per biomass that affect the disposal of ammonium oxidizers and nitrite oxidizers. A proper selection of operating conditions such as the inlet ammonium concentration and dilution rate is of great importance for the adequate design and operation of the process. The choice of these parameters also depends on the kinetic parameters. The combination of operating parameters and the kinetic parameters are important for the performance of the bioreactor (Pollice et al., 2002).

2. Process model

We consider the following model for SHARON process that was developed and validated experimentally by Volcke et al. (2006). The biological system is composed of four components. The total ammonium with concentration X_1, the total nitrite X_2, the ammonium oxidizers X_3 and the nitrite oxidizers X_4. The model assumes that the feed stream does not contain any ammonium oxidizer or nitrite oxidizer. The balance equations for the different species are given by:

$$\frac{dX_1}{dt} = U_0(U_1 - X_1) - a\mu_1(X_1, X_2)X_3 - b\mu_2(X_2)X_4 \quad (1)$$

$$\frac{dX_2}{dt} = -U_0X_2 + c\mu_1(X_1, X_2)X_3 - d\mu_2(X_2)X_4 \quad (2)$$

$$\frac{dX_3}{dt} = -U_0X_3 + \mu_1(X_1, X_2)X_3 \quad (3)$$

$$\frac{dX_4}{dt} = -U_0X_4 + \mu_2(X_2)X_4 \quad (4)$$

U_0 is the dilution rate (inverse of residence time), U_1 is the feed of total ammonium, and *a, b, c, and d* are positive stoichiometric constants. The terms μ_1 and μ_2 represent, respectively, the growth rates of ammonium oxidizers and nitrite oxidizers. They are assumed to be given by the following expressions:

$$\mu_1(X_1, X_2) = a_1\frac{c_1X_1}{(b_1 + X_1)(c_1 + X_2)} \quad (5)$$

$$\mu_2(X_2) = a_2\frac{X_2}{b_2 + X_2} \quad (6)$$

The growth rate μ_1 assumes that ammonium oxidation is inhibited by nitrite through the term c_1. When the term c_1 is very large, the inhibition by nitrite does not exist anymore. The growth rate μ_2, on the other hand, indicates that ammonium limitation is not considered. This is a simplification that was justified experimentally by Volcke (2006). The terms a_1 and a_2 represent, on the other hand, the maximum growth rate of ammonium oxidizers and nitrite oxidizers.

The model (Eqs. (1)–(6)) is rendered dimensionless using the following variables

$$\overline{X_1} = \frac{X_1}{b_1}, \quad \overline{X_2} = \frac{X_2}{c_1}, \quad \overline{X_3} = a\frac{X_3}{b_1}, \quad \overline{X_4} = b\frac{X_4}{b_1},$$
$$\bar{t} = ta_1, \quad \overline{U_0} = \frac{U_0}{a_1}\overline{\mu_1} = \frac{\mu_1}{a_1}, \quad \overline{\mu_2} = \frac{\mu_2}{a_2}, \quad \lambda_1 = \frac{b_2}{c_1},$$
$$\lambda_2 = \frac{a_2}{a_1}, \quad \lambda_3 = \frac{cb_1}{ac_1}, \quad \lambda_4 = \frac{da_2b_1}{a_1c_1b}$$

The model (Eqs. (1)–(4)) in dimensionless form therefore becomes:

$$\frac{d\overline{X_1}}{dt} = \overline{U_0}(\overline{U_1} - \overline{X_1}) - \overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} - \lambda_2\overline{\mu_2}(\overline{X_2})\overline{X_4} \tag{7}$$
$$\frac{d\overline{X_2}}{dt} = -\overline{U_0X_2} + \lambda_3\overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} - \lambda_4\overline{\mu_2}(\overline{X_2})\overline{X_4} \tag{8}$$
$$\frac{d\overline{X_3}}{dt} = -\overline{U_0X_3} + \overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} \tag{9}$$
$$\frac{d\overline{X_4}}{dt} = -\overline{U_0X_4} + \lambda_4\overline{\mu_2}(\overline{X_2})\overline{X_4} \tag{10}$$

The dimensionless specific growth rates are:

$$\overline{\mu_1}(\overline{X_1}, \overline{X_2}) = \frac{\overline{X_1}}{(1 + \overline{X_1})(1 + \overline{X_2})} \tag{11}$$
$$\overline{\mu_2}(\overline{X_2}) = \frac{\overline{X_2}}{\lambda_1 + \overline{X_2}} \tag{12}$$

The model contains several kinetic and operating parameters. The kinetic parameters are λ_1, λ_2, λ_3, and λ_4. The operating parameters are the dimensionless dilution rate ($\overline{U_0}$ and the ammonium dimensionless feed concentration ($\overline{U_1}$).

In the following, we carry out the analysis of the model.

3. Results and discussion

3.1. Analysis of the steady state behavior

The steady state solutions of the model are obtained when the left hand sides of Eqs. (7)–(10) are set to zero. In this case, the equations are equivalent, after some manipulations, to:

$$0 = \overline{U_0}(\overline{U_1} - \overline{X_1}) - \overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} - \lambda_2\overline{\mu_2}(\overline{X_2})\overline{X_4} \tag{13}$$
$$0 = -\overline{U_0X_2} + \lambda_3\overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} - \lambda_4\overline{\mu_2}(\overline{X_2})\overline{X_4} \tag{14}$$
$$0 = \overline{X_3}(-\overline{U_0} + \overline{\mu_1}(\overline{X_1}, \overline{X_2})) \tag{15}$$
$$0 = \overline{X_4}(-\overline{U_0} + \lambda_4\overline{\mu_2}(\overline{X_2})) \tag{16}$$

The analysis of Eqs. (15) and (16) reveals the following solutions:

1. $\overline{X_3} = 0$ and $\overline{X_4} = 0$.
2. $\overline{X_3} = 0$ and $\overline{X_4} \neq 0$.
3. $\overline{X_3} \neq 0$ and $\overline{X_4} = 0$.
4. $\overline{X_3} \neq 0$ and $\overline{X_4} \neq 0$.

In the following we examine each of these solutions.

For the first solution (1), substituting for the zero values of $\overline{X_3}$ and $\overline{X_4}$ into Eqs. (13) and (14) yields also $\overline{X_1} = \overline{U_1}$ and $\overline{X_1} = 0$. This solution corresponds therefore to total washout solution where all the species are washed out from the system.

The second solution (2) is not physically realistic. Since (X_4) is obtained as result of reaction of (X_3) it is therefore impossible that (X_4) exists when (X_3) does not.

For the third solution (3), when $\overline{X_3} \neq 0$, Eq. (15) leads to

$$\overline{U_0} = \overline{\mu_1}(\overline{X_1}, \overline{X_2}) \tag{17}$$

Substituting $\overline{X_4} = 0$ in Eqs. (13) and (14) yields

$$0 = \overline{U_0}(\overline{U_1} - \overline{X_1}) - \overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} \tag{18}$$
$$0 = -\overline{U_0X_2} + \lambda_3\overline{\mu_1}(\overline{X_1}, \overline{X_2})\overline{X_3} \tag{19}$$

Solving these two equations yields,

$$\overline{U_0X_2} = \lambda_3(\overline{U_1} - \overline{X_1}) \tag{20}$$
$$\overline{X_3} = (\overline{U_1} - \overline{X_1}) \tag{21}$$

From Eqs. (20) and (21) it can be seen that both X_2 and X_3 are physically realistic (i.e., positive) given that $\overline{U_1}$ is always larger than $\overline{X_1}$. Substituting for the expression of $\overline{X_2}$ (Eq. (20)) and the expression of growth rate $\overline{\mu_1}$ (Eq. (11)) into Eq. (13) yields the quadratic equation for $\overline{X_1}$:

$$b\overline{X_1}^2 + c\overline{X_1} + d = 0 \tag{22}$$

with

$$b = \lambda_3$$
$$c = -1 - \lambda_3 - \overline{U_0} - \lambda_3\overline{U_1}$$
$$d = -\overline{U_0} - \lambda_3\overline{U_1}$$

The real solution(s) of this quadratic equation are substituted into Eqs. (13) and (14) to yield the expressions for $\overline{X_2}$ and $\overline{X_3}$

For the solution (4), substituting in Eq. (15) yields

$$\overline{U_0} = \overline{\mu_1}(\overline{X_1}, \overline{X_2}) \tag{23}$$

and

$$\overline{U_0} = \lambda_4\overline{\mu_2}(\overline{X_2}) \tag{24}$$

Substituting the expression of the growth rate $\overline{\mu_2}$ yields the following simple equation,

$$b\overline{X_2} + c = 0 \tag{25}$$

with

$$b = \overline{U_0} - \lambda_4$$
$$c = \overline{U_0}\lambda_1$$

As to Eq. (23), it is equivalent to

$$\overline{U_0} = \frac{\overline{X_1}}{1 + \overline{X_1}}\frac{1}{1 + \overline{X_2}}$$

Expanding this equation yield another simple equation

$$b\overline{X_1} + c = 0 \tag{26}$$

with

$$b = \overline{U_0} - \frac{1}{1 + \overline{X_2}}$$
$$c = \overline{U_0}$$

To obtain the values of the state variables for this case, we proceed as follows: We first solve (Eq. (25)) for $\overline{X_2}$. The solution of $\overline{X_2}$ is substituted into the simple Eq. (26) to solve for $\overline{X_1}$. Substituting for the calculated $\overline{X_1}$ and $\overline{X_2}$ into the Eqs. (13) and (14) we can solve for $\overline{X_3}$ and $\overline{X_4}$.

When analyzing the behavior of the process we need to define some performance criteria. The following criteria are used:1. The process efficiency. It can be defined by

$$E_f = \frac{X_1}{U_1} \quad \text{or} \quad E_f = \frac{X_2}{U_1}$$

These criteria define the amounts of X_1 or X_2 in the bioreactor. Smaller values of E_f mean better biodegradation. In dimensionless term, it becomes,

$$\overline{E_f} = \frac{\overline{X_1}}{\overline{U_1}} \quad \text{or} \quad \overline{E_f} = \frac{\overline{X_2}}{\overline{U_1}}$$

2. Process productivity: It can be defined by

$$P_r = U_0 X_3 \quad \text{or} P_r = U_0 X_4$$

This performance defines the amount of (X_3) or (X_4) produced. In dimensionless terms this criteria becomes:

$$\overline{P_r} = \overline{U_0 X_3} \quad \text{or} \quad \overline{P_r} = \overline{U_0 X_4}$$

The numerical analysis of the model (7)–(10) is carried out using the nominal values shown in Table 1. The corresponding dimensionless variables are shown in Table 2.

Fig. 1 shows the variations of the ammonium concentration with the dilution rate, for the nominal values of Table 2. The three steady state solutions discussed previously are found in this figure. The upper horizontal line (CDE) is the line of total washout. The line (ABC) represents the line of the washout of nitrite oxidizers while the line (AB) is the line of the coexistence of the different species. Therefore, operating the bioreactor for dilution rates larger than point (C i.e., $\overline{U_0} = 0.9475$) will lead to total washout of all species. Dilution rates between (B i.e., $\overline{U_0} = 0.446$) and (C) lead to the washout of the nitrite oxidizers, since the other branch (dash) is unstable. Choosing, on the other hand, dilution rates between A and B will lead to the coexistence of the species, since the other two branches are unstable. We conclude therefore that it is region (BC) which offers the most interesting feature, since in this region the nitrite oxidizers are washed out and therefore we can prevent the oxidation of nitrite to nitrate.

Table 1 Nominal values of model parameters.

Parameter	Value
a_1	2.1 (day^{-1})
b_1	4.73
c_1	837
a_2	1.05 (day^{-1})
b_2	109
c_2	0.01
d_2	1000
e_2	1000
a	16
b	0.2
c	58.6
d	15.8

Table 2 Dimensionless model parameters.

Parameter	Value
λ_1	0.1302
λ_2	0.5
λ_3	0.215
λ_4	0.223
$\overline{U_0}$	2.1
$\overline{U_1}$	15

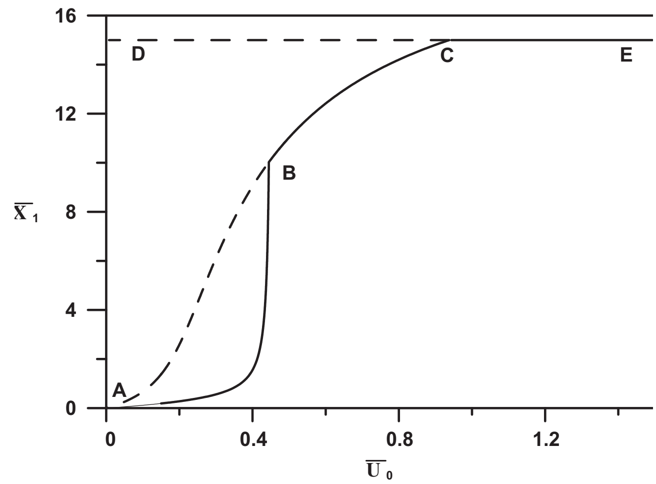

Figure 1 Variations of dimensionless ammonium concentration with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable branch.

Fig. 2 shows the variations of the nitrite concentration for the same conditions of Fig. 1. The same regions of Fig. 1 are also found in this region. It can also be seen that the nitrite concentration increases with dilution rate throughout the region of coexistence (AB). It then decreases throughout the region (BD) where the nitrite oxidizers are washed out, before being itself washed out in the total washout line (DE).

Fig. 3 shows, on the other hand, the concentration of ammonium oxidizers. It can be seen that in the region of coexistence (AB), the concentration of the ammonium oxidizers is almost constant for dilution rates up to 0.4. It then decreases sharply until it reaches point (B). On the region (BC), the concentration of ammonium oxidizers decrease smoothly until it reaches region (C) where all species are washed out from the bioreactor.

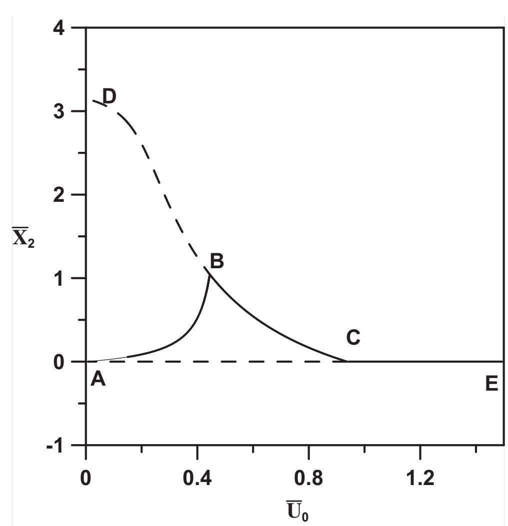

Figure 2 Variations of dimensionless nitrite concentration and ammonium concentration with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable branch.

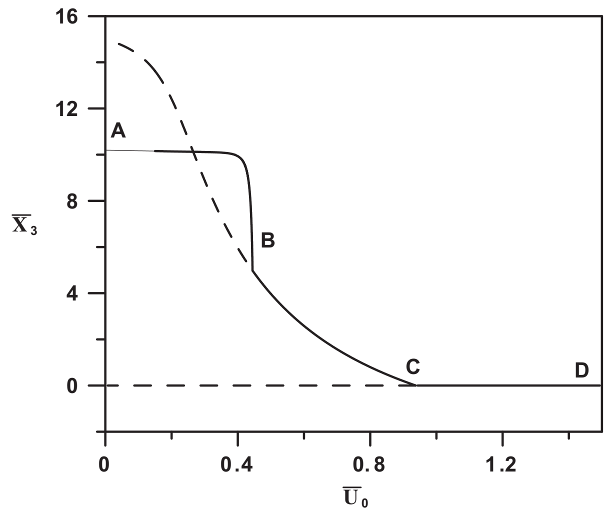

Figure 3 Variations of dimensionless ammonium oxidizers concentration with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable branch.

Fig. 4 shows the variations of nitrite oxidizers. It can be seen that the concentration decreases sharply throughout the region (AB) of coexistence. At point (B), the nitrite oxidizers are washed out. It should be noted that in this figure, the total washout line (where all species are washed out) and the washout line of the nitrite oxidizers alone are all lumped in one single branch.

3.2. Construction of operating diagrams

The analysis, shown in Figs. 1–4, illustrates that there is a critical region of dilution rates where nitrite oxidizers are washed out. Therefore it is possible, if the bioreactor is operated in this region, to prevent further oxidation of nitrite to nitrate. This critical region (BC) in Figs. 1 and 2 is therefore important. The limits of this region are point (B) and (C). Point (B) corresponds to the crossing of the region (BC) with the region of coexistence (AB). Point (C), on the other hand, corresponds to the crossing of region (AB) with the total washout.

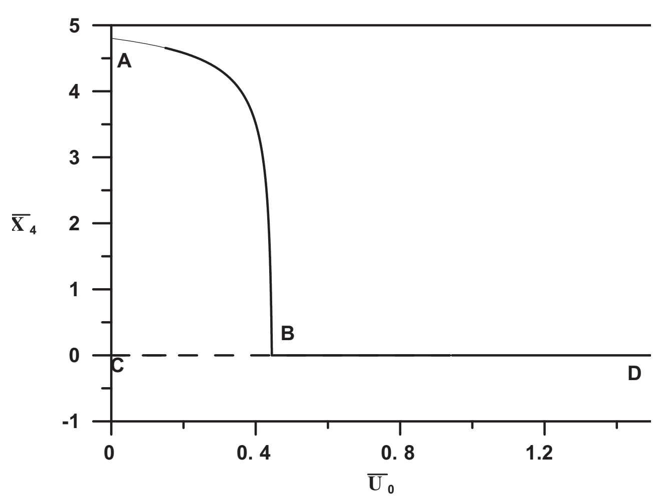

Figure 4 Variations of dimensionless ammonium oxidizers concentration with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable branch.

In the following section, we construct practical diagrams that show the effect of the bioreactor kinetic and operating parameters on the extent of this region.Fig. 5 shows the effect of the dimensionless feed concentration of ammonium on the domains of system behavior. The line denoted by (B) shows the variations of point (B) of Figs. 1 and 2 while the same holds for point (C). The two curves in Fig. 5 separate the domain $(\overline{U_1}, \overline{U_0})$ in three regions. In the upper region (III), the system is washed out completely. In the lower region (I), there is the coexistence of species. The region (II) is the region of interest, and corresponds to the washout of nitrite oxidizers. It can be seen from the diagram that increasing the feed concentration of ammonium tends to increase the region (II), since the branch (C) increases while the branch (B) increases for small values of the feed concentration but saturates at higher values.

Figs. 6–8 show the effect of the kinetic parameters. Fig. 6 shows the effect of λ_1. It can be seen that branch (C) is insen-

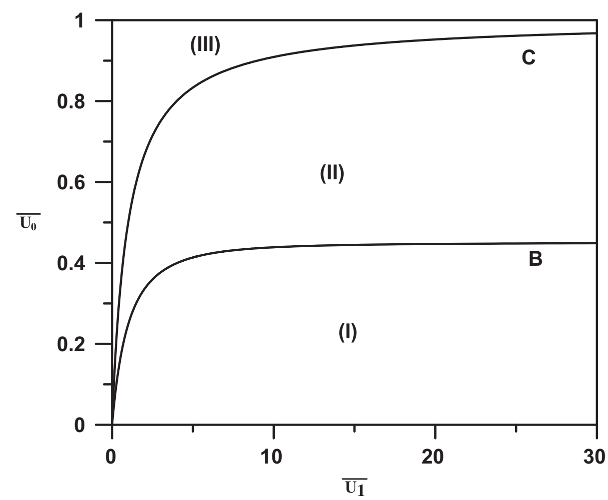

Figure 5 Domains of the coexistence and washout in the parameter space $(\overline{U_1}, \overline{U_0})$.

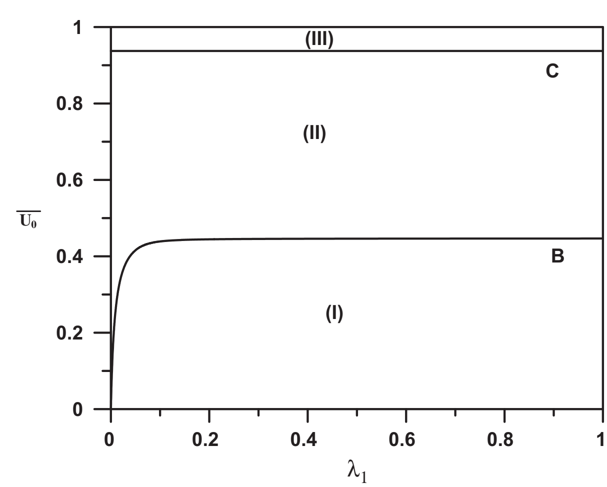

Figure 6 Domains of the coexistence and washout in the parameter space $(\lambda_1, \overline{U_0})$.

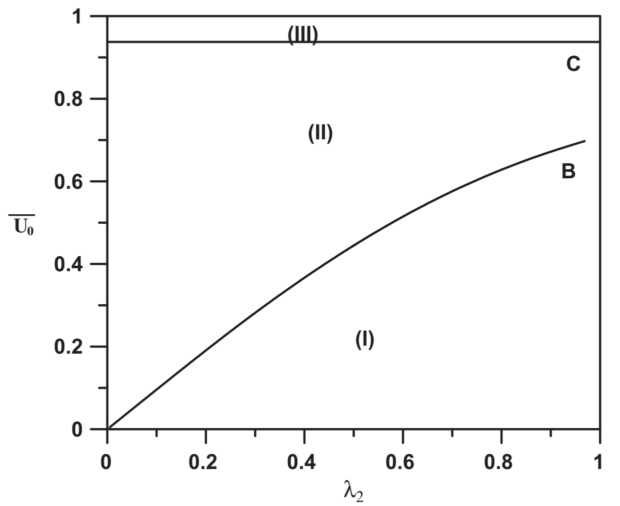

Figure 7 Domains of the coexistence and washout in the parameter space ($\lambda_2, \overline{U_0}$).

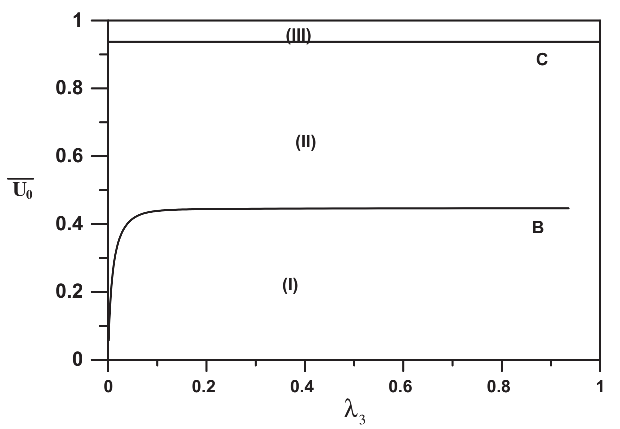

Figure 8 Domains of the coexistence and washout in the parameter space ($\lambda_3, \overline{U_0}$).

sitive to variations in λ_1 while the branch (B) increases fast for small values of λ_1 but saturates at high values. Therefore the region (II) is practically invariable for larger values of λ_1 but the region increases for smaller values. The effect of λ_2 is shown in Fig. 7. As it can be seen while the branch (C) is unchanged, the increase in λ_2 has strong effects, as the region (II) narrows at large values of this kinetic parameter. Finally, the effect of the other kinetic parameter λ_3 is shown in Fig. 8. This effect is almost similar to the effect of λ_1, shown in Fig. 6.

3.3. Performance of the system

Next we examine the performance of the bioreactor. The performance is shown only for nontrivial conditions. In this way, the total washout line is excluded, as well as the productivity for nitrite oxidizers when they are washed out.

Fig. 9 shows variations of the efficiency of ammonium with the dilution rate. It can be seen that the efficiency follows the same trend as that of Fig. 1. Fig. 10 shows also the efficiency for nitrite. Again the figure shows similar trends as Fig. 2.

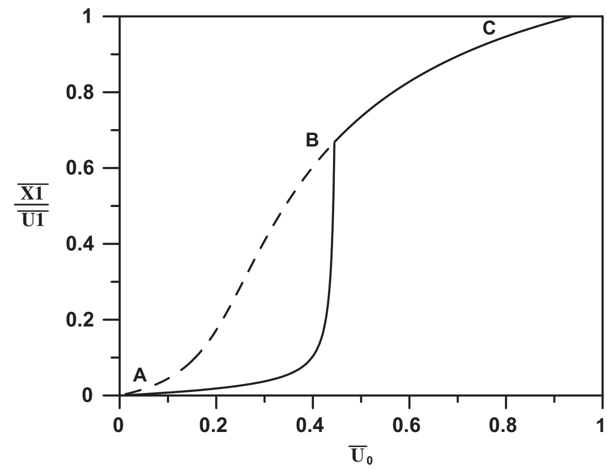

Figure 9 Variations of efficiency with the dimensionless dilution rate for. Solid line, stable branch; dash line, unstable.

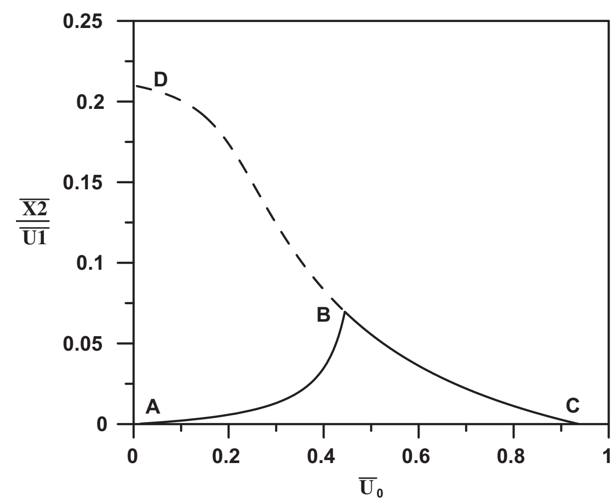

Figure 10 Variations of efficiency with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable.

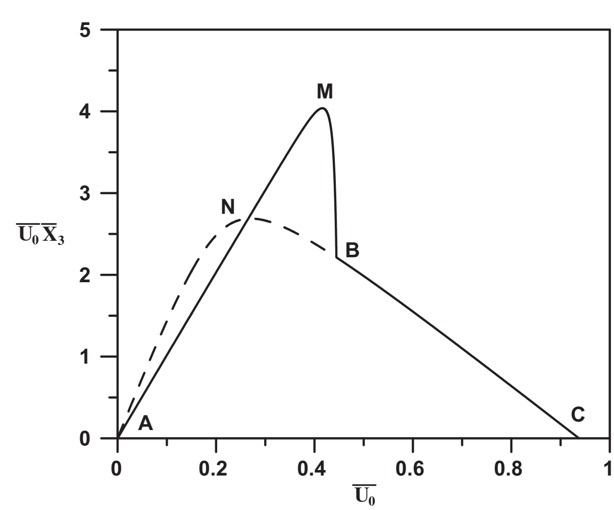

Figure 11 Variations of productivity with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable.

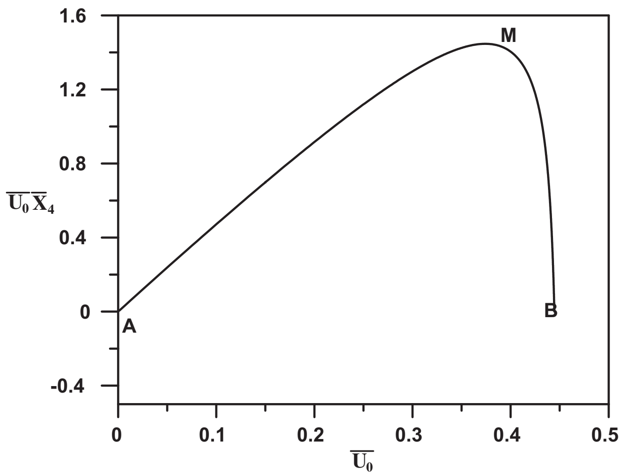

Figure 12 Variations of productivity with the dimensionless dilution rate. Solid line, stable branch; dash line, unstable.

The variations of the productivity show, on the other hand, an interesting trend. Fig. 11 shows that the line (AB) of coexistence shows a maximum at point (M). The line (ABC) of nitrite oxidizers washout also shows a maximum point (N) at an unstable branch. Since we are interested in operating the bioreactor in the region (ABC), it can be seen that a feedback control is needed to stabilize the operation of the bioreactor at the point (N) of maximum productivity.

Fig. 12 shows, on the other hand, the productivity for nitrite oxidizers. In this plot we show only the region of coexistence, since the other regions (total washout or nitrite oxidizers washout) yield zero values for the productivity. It can be seen from this figure that the productivity also reaches a maximum at a stable point.

4. Conclusions

In this paper, the static behavior of a validated model for a nitritation process under time invariant conditions were analyzed. The following conclusions can be obtained:

- The analysis of the unforced system showed that there is effectively a critical region of dilution rates for which the oxidation of nitrite to nitrate can be prevented.
- This critical region can be increased by an increase in the ammonium feed concentration and is also sensitive to the ratio of maximum specific growth rates of ammonium oxidizers to nitrite oxidizers.
- The productivity of the unforced reactor in the critical region reaches a maximum but in an unstable region of dilution rate. A feedback control is needed if the operation is desired in this region.

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