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Kinetics of cross-inhibited denitrification of a high load wastewater

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Abstract

Batch denitrification of a synthetic saline medium at 37 °C, pH 7.5 was studied. A kinetic expression and an inhibition function which adequately expresses inhibition and cross-inhibition by nitrate and nitrite in denitratation and denitritation steps occurring in high organic and nitrate load wastewater treatment was determined. Denitratation and denitritation rates were measured independently. Monod- and Haldane-type kinetics were used for denitratation and denitritation, respectively. The maximum specific rate, $k_{NO_3} = 12.7$ mg NO_3-N/g VSS h, and the saturation constant, $K_{S_{NO_3}} = 0.47$ mg NO_3-N/L were calculated for denitratation. The maximum specific rate, $k_{NO_2} = 13.8$ mg NO_2-N/g VSS h, the saturation constant $K_{S_{NO_2}} = 0.36$ mg NO_2-N/L and the inhibition constant $K_{I\text{-}NO_2} = 906$ mg NO_2-N/L were calculated for denitritation. Among other functional forms the Levenspiel inhibition model for denitratation and denitritation kinetics was proposed. The Levenspiel constants were $S_{NO_2\text{-}M} = 1149.4$ mg NO_2-N/L, $S_{NO_3\text{-}M} = 34.7$ mg NO_3-N/L, $\beta_1 = 1.578$ and $\beta_2 = 1.005$, respectively. The fitted experimental values show a good representation of the denitrification of effluents with high nitrogen load containing both nitrate and nitrite. Parametric sensitivity analysis show that inhibition functions (f_i) and dimensionless constants (β_i) greatly affect predicted nitrite and nitrate consumption rates.
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Keywords: Denitrification; Denitratation; Denitritation; Kinetics; Saline medium

1. Introduction

Wastewaters from salmon and other fish processes feature high salt, proteins, nitrate and nitrite concentrations [1,2], which are treated stepwise anaerobically and aerobically to reduce organic matter, producing nitrate and nitrite. Often, resulting nitrite and nitrate concentrations exceed levels imposed by environmental legislation. In this context, biological denitrification could be used to reduce nitrogen oxides to acceptable levels. Denitrification is a process of anoxic microbial respiration in which nitrate is reduced to molecular nitrogen with nitrite as an intermediate product [3]. The electron donor is generally organic matter, which may be obtained from the treated wastewater [4]. Denitrification systems could be used to treat anaerobically treated fishery wastewaters. Such effluents contain significant volatile fatty acids loads [5], which are known electron donors for denitrification [6]. Denitrification includes two major biological processes, namely, denitratation (i.e. nitrate reduction to nitrite) and denitritation (i.e. nitrite reduction to gaseous N_2). Moreover, nitrate and nitrite have been found to inhibit denitrification systems [3,7–13]. Although detailed information is provided in the literature for the inhibition on each individual denitrification step, little attention has been paid to possible cross-inhibition of reactants.

The objective of the present study is to experimentally analyze the inhibitory effects of nitrate and nitrite, at high organic and nitrate load and saline conditions. Denitratation and denitritation rates are assessed separately in order to determine inhibition and cross-inhibition by nitrate and nitrite. A suitable inhibition model is proposed.

1.1. Mathematical modeling

Denitrification processes involve the initial nitrate (NO_3^-) reduction to nitrite (NO_2^-), followed by further reduction to nitric oxide (NO), nitrous oxide (N_2O) and finally molecular nitrogen (N_2). Although denitratation and denitratation are complex biological processes, the overall denitrification pathway from nitrate to gaseous nitrogen may be simplified as illustrated in Fig. 1. Based on experimental findings, nitrite and nitrate inhibition mechanisms proposed in this work are included in this figure.

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Nomenclature

f_1	inhibition function on denitratation for nitrite
f_2	inhibition function on denitritation for nitrate
i	subunit
$K_{\text{I-NO}_2}$	Haldane inhibition constant in denitritation (mg NO_2-N/L)
k_{NO_2}	maximum specific nitrite reduction rate (mg NO_2-N/g VSS h)
k_{NO_3}	maximum specific nitrate reduction rate (mg NO_3-N/g VSS h)
$K_{S_{NO_2}}$	half-saturation constant for nitrite (mg NO_2-N/L)
$K_{S_{NO_3}}$	half-saturation constant for nitrate (mg NO_3-N/L)
r_{NO_2}	specific nitrite reduction rate (mg NO_2-N/g VSS h)
r_{NO_3}	specific nitrate reduction rate (mg NO_3-N/g VSS h)
$r^*_{NO_2}$	reference specific nitrite reduction rate (mg NO_2-N/g VSS h)
$r^*_{NO_3}$	reference specific nitrate reduction rate (mg NO_3-N/g VSS h)
$S_{\text{I-NO}_x}$	non-competitive inhibition constant (mg NO_x-N/L)
$S_{\text{NO}_x\text{-M}}$	maximum concentration at which specific reduction rate is zero (mg NO_x-N/L)
S_{NO_x}	nitrate or nitrite concentration (mg NO_x-N/L)
X	biomass concentration (g VSS/L)

Greek symbols

β_i	inhibition dimensionless constant for Eq. (3)
γ_i	inhibition dimensionless constant for Eq. (4)

Several attempts have been made to model the inhibition of both steps. Nitrate reduction has usually been described by a Monod-type kinetics which means substrate limiting, but no substrate inhibition [7,14,15]. Several authors have demonstrated nitrite inhibition in the denitratation stage [7,8,16]; affecting microbial growth. These experimental evidences should be included in the denitratation kinetics.

On the other hand, Haldane kinetics, representing substrate inhibition, has been proposed for nitrite reduction [10,13,17]. Almeida et al. [7] and Wang et al. [13] proposed a nitrate inhibition model in denitritation for *Pseudomonas fluorescens* and *P. denitrificans*, respectively. These models represent adequately the processes, but each step – denitratation and denitritation – are separately studied. Glass and Silverstein [10] established the existence of two different maximum denitritation rates, one in the presence of nitrate (k'_2), and a second one (k_2) once the nitrate has been completely reduced. Huang et al. [11] presented a "distributed fraction of nitrate-reductase enzyme" (f), which is an empirical solution similar to the Glass and Silverstein [10] approach. Finally, Schönharting et al. [12] applied a "two-site non-competitive" model without showing experimental kinetic parameter evaluation.

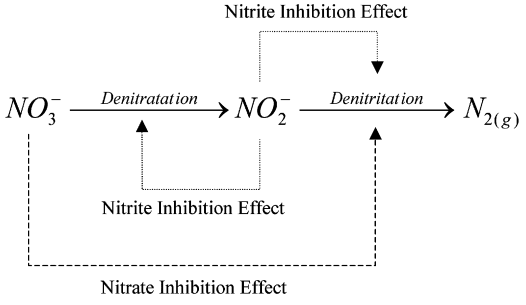

Fig. 1. Denitrification process with inhibition of denitritation and denitratation steps.

Our preliminary work showed that these models represent very well nitrite and nitrate consumption at low concentrations. Therefore, this work proposes the following integrated model for the combined denitratation and denitratation of species in a wide range of concentrations:

$$r_{NO_3} = \left[\frac{k_{NO_3} S_{NO_3}}{K_{S_{NO_3}} + S_{NO_3}}\right] f_1 \quad (1)$$

$$r_{NO_2} = \left[\frac{k_{NO_2} S_{NO_2}}{K_{S_{NO_2}} + S_{NO_2} + S^2_{NO_2}/K_{\text{I-NO}_2}}\right] f_2 \quad (2)$$

where f_1 and f_2 represent the inhibition functions of nitrite on denitratation, and of nitrate on the denitritation kinetics, respectively. According to the previously cited literature, Monod and Haldane kinetic models were used for denitratation and denitritation kinetics, respectively. Thus, this work will focus on the mathematical corrections that the inhibition of species poses on these models. The following inhibitory models were used in this work [18–20]:

- Levenspiel's model:

$$f_i = \left(1 - \frac{S_{NO_x}}{S_{\text{NO}_x\text{-M}}}\right)^{\beta_i} \quad (3)$$

- Luong's model:

$$f_i = 1 - \left(\frac{S_{NO_x}}{S_{\text{NO}_x\text{-M}}}\right)^{\gamma_i} \quad (4)$$

- Non-competitive model:

$$f_i = \left(\frac{S_{\text{I-NO}_x}}{S_{\text{I-NO}_x} + S_{NO_x}}\right) \quad (5)$$

In Eqs. (3)–(5) when $i = 1$, NO_x is NO_2; when $i = 2$, NO_x is NO_3.

Most of the kinetic models have an empirical nature and are based on models describing enzymatic inhibition. Luong's model [18] is a modification of the general model proposed by Levenspiel [19], while the non-competitive model constitutes an extension of the non-competitive inhibition of enzymatic reactions [20]. The goal of these models is to correct the rate of substrate consumption by a factor that depends on the inhibitor concentration; however, due to their simplicity, the previous models present some limitations. The non-competitive model, although based on reaction enzyme mechanisms has the

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