Chemical Engineering Journal 268 (2015) 300–310

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

# Model based predictive control for energy efficient biological nitrification process with minimal nitrous oxide production ☆

Chitta Ranjan Behera [a], Babji Srinivasan [a,*], Kartik Chandran [b], Venkat Venkatasubramanian [c]

[a] Department of Chemical Engineering, IIT Gandhinagar, VGEC Campus, Chandkheda, Ahmedabad 382424, India
[b] Department of Earth and Environmental Engineering, Columbia University, New York 10027, USA
[c] Department of Chemical Engineering, Columbia University, New York 10027, USA

## HIGHLIGHTS

- Experiment based model for $N_2O$ production by ammonium oxidizing bacteria.
- Nonlinear soft sensing strategies used to estimate all variables under uncertainties.
- Model based control strategy for optimal aeration profile with minimal $N_2O$ production.
- Method is fairly insensitive to parameter variations in the model.

## ARTICLE INFO

*Article history:*
Received 16 October 2014
Received in revised form 7 January 2015
Accepted 11 January 2015
Available online 22 January 2015

*Keywords:*
Biological nitrogen removal
Nitrous oxide emission
Extended Kalman filter
Nonlinear model predictive control

## ABSTRACT

Recent studies reveal that Ammonium Oxidizing Bacteria (AOB) in the Biological Nitrification Removal (BNR) process is one of the main contributors for Nitrous Oxide ($N_2O$) emissions, a powerful greenhouse gas having a potential of 300 times that of Carbon Dioxide ($CO_2$) (IPCC, 2011; Ravishankara et al., 2009 [1,2]). Though a few models have been proposed to understand the behaviour of $N_2O$ production by AOB under various conditions, there exists hardly any work that aim towards development of a control strategy that utilizes these kind of models to mitigate $N_2O$ production. In this work, a model is developed based on the experimental studies (Ni et al., 2013 [3]) that capture the dynamics of $N_2O$ during recovery to aerobic conditions, after a period of anoxia, a common practice in nitrogen removal process. Subsequently, this model is employed in soft sensing using Extended Kalman Filter (EKF) to estimate $N_2O$ concentration and develop an advanced model based control strategy for energy efficient BNR process with minimal $N_2O$ production. This control strategy provides an aeration profile that minimizes $N_2O$ production and energy consumption (less pumping cost) apart from meeting the desired ammonium level at the output. The key features of the proposed model based control strategy are: (i) only continuous measurements of DO is required and, (ii) fairly insensitive to fluctuations in the influent ammonium loading and changes in the model parameters.



## 1. Introduction

Accumulation of nitrogen compounds (ammonia, nitrate, nitrite, etc.) in surface water creates serious ecological problems for aquatic life as well as humans. There are several major effects linked to the discharge of nitrogen compounds into water streams (e.g., rivers, lakes) such as eutrophication, fish kills and nitrate contamination of ground water leading to Blue Baby Syndrome [1,4]. These consequences coupled with an ever increasing need for drinking water demands development of a technology to treat waste water from industry (Fertilizer, Dairy, Oil Refinery, etc.) or household in an eco-friendly fashion. Biological Nitrogen Removal (BNR) is a commonly used process to maintain the level of total nitrogen (TN) below 3 (mg-N/l) [5] in the treated effluent. In the BNR process, $NH_4^+$ in the influent is oxidized under aerated conditions to $NO_2^-$ using autotrophic Ammonium Oxidizing Bacteria (AOB) which is subsequently oxidized to $NO_3^-$ by nitrification bacteria. However, this step is avoided by performing a nitrification with low Dissolved Oxygen (DO) to eliminate the action of nitrifying bacteria. This process is followed by reduction of nitrites to $N_2$ gas under anoxic conditions using a set of heterotrophic denitrifying bacteria.

☆ This work was partly supported by funds from Department of Science and Technology Fast track program SERB/ET-26/2013, Government of India.

* Corresponding author.
*E-mail address:* babji.srinivasan@iitgn.ac.in (B. Srinivasan).

http://dx.doi.org/10.1016/j.cej.2015.01.044
1385-8947/

Recent studies have identified that AOB produces $N_2O$ during nitrification and autotrophic denitrification with the majority of production occurring in the aerobic zones of the BNR process [6,7]. A long term study carried out on municipality Waste Water Treatment Plant (WWTP) at Kralingseveer reveal that nitrous oxide alone (greenhouse gas measurements include $CO_2$, $N_2O$ and $CH_4$) contributed to around 78.4% of total greenhouse gas footprint, expressed as $CO_2$ equivalent [8]. Both nitrification and denitrification processes produce significant amount of $N_2O$; however it has been shown that AOB involved in the nitrification process can produce $N_2O$ at a higher rate [9,10]. A detailed study of nitrous oxide production from different types of waste water treatment presented in [11,12] show that $N_2O$ emission varies in the range of 0.001–25% of the total nitrogen load which is related to its production. It is observed that $N_2O$ formation is high during recovery from oxygen limitation/anoxia to aerobic state with large ammonium loading and/or excess aeration [10].

Several works have been performed to understand the mechanisms and changes in the electron donor concentration leading to production of $N_2O$ under different conditions of aeration [13–16]. Two main pathways that are found to produce nitrous oxide are: (i) HAO (HydroxylAmine Oxidoreductase) mediated aerobic hydroxylamine reduction and, (ii) NIR (Nitrite Reductase) and NOR (Nitric Oxide Reductase) mediated nitrifier denitrification [14,13]. Changes in the concentration of electron donors under various conditions in the BNR plant influence these pathways resulting in production of nitrous oxide. Recently a detailed Stoichiometric Metabolic Network (SMN) network model has been developed to understand the metabolism of *Nitrosomonas europaea* (representative AOB) [17]. It has been shown that $N_2O$ is primarily produced due to the difference in production and consumption of electrons when subjected to aerated-anoxic-aerated transitions. Unlike this detailed SMN model, several models (relatively simple compared to SMN) based on nitrifier denitrification pathways of AOB has been developed to capture the dynamics of $N_2O$ production. However, as discussed in [3] not a single model is able to mimic the entire observed experimental results. Given the fact that the underlying mechanisms of $N_2O$ production from AOB is in nascent stages, there are only a few investigations performed to understand the impact of carbon source and oxygen supply on the overall $N_2O$ production [18–21]. In addition, literature studies indicate that systematic study pertaining to development of control strategy for energy efficient BNR process with focus on $N_2O$ production is largely uncharted.

The objective of this study is to develop an advanced model based predictive control strategy by utilizing the available knowledge in the behaviour of AOB for an eco-friendly energy efficient BNR process. To accomplish this goal, a nitrifier denitrification pathway model for AOB is developed based on lab-scale experiments with pure cultures of *Nitrosomonas europaea* [16]. The primary focus is specifically on ammonia oxidation for this work, since autotrophic ammonia oxidizing bacteria (AOB) can produce $N_2O$ but cannot reduce it any further. Owing to this limitation, it is now being recognized that AOB could potentially be worse contributors to $N_2O$ emissions from BNR processes than heterotrophic denitrification [10]. Therefore, this work represents a first step towards the focused modeling and control of $N_2O$ production, a powerful greenhouse gas [2], from possibly the higher contributor in BNR processes. It is expected that this work would be able to potentially integrate as a module into future plant-wide modeling and control efforts on a wide range of BNR systems (which could be quite varied among themselves).

To establish the Model Predictive Control (MPC) strategy, it is important to have an online knowledge of all the state variables in the developed mechanistic model. However, it is difficult/some times impossible to measure several variables online due to the cost and technology involved. For instance, online measurement of cell concentration, nitrous oxide and nitrites are difficult and expensive. Therefore, soft sensing of these state variables are performed using an Extended Kalman Filter (EKF) approach with dissolved oxygen as the only continuously measured variable along with the knowledge of influent ammonium loading. EKF is a model based soft sensing method used to estimate the unmeasurable states; given a description of the relation between state variables and the noises affecting these states. For linear systems, it has been shown that Kalman filter is the best optimal method to estimate the unmeasured states [22,23]. However for nonlinear systems, EKF is a widely used de facto approach which linearizes the nonlinear system about current mean and covariance by using a Taylor series and then estimates the unmeasured states by a prediction and correction approach [24,25]. This technique does not guarantee optimal estimation as there is loss of information during the linearization process [26,27] and performs poorly when presented with constraints [28]. Nevertheless, EKF is known to provide convergent results for varying degrees of parameter changes in the model and also known as de facto filter [27,29,25]. The advantages of using EKF in the proposed strategy are: (i) provide an estimate of all the variables in the process at any time with noisy DO measurements and, (ii) fairly insensitive to mismatch between the true process and model. MPC utilizes these estimated states and solves a multi-objective optimization problem of minimizing energy utilization and $N_2O$ production using the developed nonlinear model. Sensitivity analysis of the proposed control strategy to several key parameters in the model are analyzed. Our results not only indicate that it is possible to achieve this objective using predictive control but also indicate that it could be accomplished with a limited process knowledge.

## 2. Materials and methods

### 2.1. BNR process – mechanistic model development & evaluation

In BNR process, typically ammonium ($NH_4^+$) is first converted to nitrite ($NO_2^-$) by the action of AOB (as shown in Eq. (1)) which is then further oxidized to nitrate ($NO_3^-$) by nitrite-oxidizing bacteria (NOB) as with the presence of dissolved oxygen (shown in Eq. (2)). Denitrification is the process of nitrate reduction into nitrite and then into molecular nitrogen by denitrifying bacteria (DB). An alternate way is to perform partial nitritation where NOB is suppressed by manipulating DO, pH and temperature [30]. Nitrite resulting from partial nitritation process is then converted to molecular nitrogen by DB. This process has the following advantages: a lower oxygen requirement during nitrification (i.e., 25% less compared to nitrogen removal over nitrate), lower organic carbon consumption in denitrification (i.e., 40% less) and a lower sludge production [31]. Also, nitrite reduction to molecular nitrogen using anaerobic ammonium oxidation (ANNAMOX) process with ammonium as electron donor, resulting in a saving of 50% of oxygen and no carbon source for reduction [32]. Considering that the number of applications of energy efficient alternates using partial nitritation is going to increase compared to that of the conventional biological nitrogen removal via nitrates, a model of the BNR process is developed using experimental data obtained from studies on ammonia oxidizing bacteria, subjected to influent ammonia-N concentrations from 28 mg-N/l to 280 mg-N/l under partial nitritation conditions. During model development, ammonium is not considered as electron donor (as in ANNAMOX process) since the main focus is on conversion of ammonium to nitrite by AOB.

$$NH_4^+ + 2HCO_3^- + 1.5O_2 \rightarrow NO_2^- + 3H_2O + 2CO_2 \quad (1)$$

$$NO_2^- + 0.5O_2 \rightarrow NO_3^- \quad (2)$$

The mechanistic model of $N_2O$ production by Autotrophic AOB is developed for a continuous reactor with the mole balance equation: