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Modelling deammonification in biofilm systems: Sensitivity and identifiability analysis as a basis for the design of experiments for parameter estimation

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Abstract

A procedure for selecting parameters for identifiability analysis and parameter estimation and for the design of experiments as a basis for parameter estimation was described for a model of deammonification in biofilm systems. A larger number of parameters were identifiable using data from batch experiments than from continuous reactor operation. However, not all sensitive parameters were identifiable from experimental data due to a large degree of parameter correlation.

Keywords: deammonification, anammox, biofilm modelling, identifiability, experimental design

1. Introduction

For the treatment of wastewater with high ammonium concentrations and low organic carbon to nitrogen ratios (C/N ratio) classical biological nitrogen elimination becomes cost-intensive. Thus, new biological nitrogen elimination processes, like deammonification (Helmer et al., 1999a) and CANON (Strous, 2000), attract increasing interest. These processes are a combination of partial oxidation of ammonium to nitrite and anaerobic ammonium oxidation (anammox). Anaerobic ammonium oxidation converts ammonium and nitrite directly to dinitrogen gas. Both steps of the deammonification can be combined in biofilms or granules into a single-stage operation (Helmer et al., 1999b; Sliekers et al., 2002).

Modelling and simulation of the deammonification can help to optimise the process. For the application of the model it is essential to estimate a set of model parameters that fit the experimental data. To be able to estimate a unique set of parameters it is necessary that the model parameters are not correlated and that the experimental data has sufficient information content with respect of the parameters that are estimated. The ability to obtain a unique parameter set that is able to describe the behaviour of a system is called "identifiability" (Petersen, 2000). Structural/theoretical identifiability is based on the model structure and deals with the possibility to obtain a unique value for each parameter from a set of noise-free (perfect) data (Dochain et al., 1995). In contrast, practical identifiability deals with the question whether the experimental and noise corrupted data available are informative enough for giving accurate parameter values (Vanrolleghem et al., 1995). The design of an experiment, e.g. mode of reactor operation, operation conditions, measured variables and measuring intervals, has an influence on the identifiability of the parameters (Holmberg, 1982; Dochain and

Vanrolleghem, 2001). Hence, designing an experiment which provides as much information as possible to determine and estimate the regarded parameters can help to reduce the experimental effort. Van Hulle (2005), for example, performed an experimental design to determine the nitrite inhibition constant for the anammox bacteria from ammonium and nitrite concentration profiles in the bulk liquid of a biofilm reactor.
In this paper, a procedure for selecting parameters for identifiability analysis and following parameter estimation as well as the design of experiments for the estimation of parameters are presented.

2. Materials and methods

2.1. Mathematical model
The model included growth and inactivation of aerobic ammonium oxidisers, X_{NH}, aerobic nitrite oxidisers, X_{NO}, and anaerobic ammonium oxidisers, X_{AN}. The growth processes were described with Monod kinetics for substrate utilisation. Ammonium consumption for biomass growth was neglected due to the low production of autotrophic biomass. Inactivation processes were defined for all three organism groups which reduced the amount of active biomass and formed inactive or inert biomass. Default values for the parameters were taken from Hao et al. (2002). The affinity constant for oxygen of the nitrite oxidisers was modified to obtain a better fit to measured data in steady state. The model describing the kinetics was integrated into the biofilm compartment of AQUASIM (Reichert, 1998) which assumes a one-dimensional biofilm structure.
The reactor set-up of the simulated continuously operated reactor was based on layout and operation parameters of a lab-scale moving-bed biofilm reactor operated for deammonification (Hippen et al., 2001). The layout of the batch reactor was according to the set-up used by Helmer et al. (2001) to investigate the conversion processes in the lab-scale moving-bed plant for deammonification. The biofilm thickness in the model was set to 450 µm as biofilm thicknesses of 420 to 510 µm were measured on the moving-bed carriers of the investigated lab-scale moving-bed reactor (Tromm, 1999).

2.2. Sensitivity and identifiability analysis
In the following, the used procedure for parameter selection and identifiability analysis is briefly described. The procedure is presented in more detail by Brockmann (2006).

2.2.1. Selection of parameters for identifiability analysis and subsequent parameter estimation
The selection of parameters for identifiability analysis and parameter estimation was based on results of a regional steady state sensitivity analysis. The regional sensitivity analysis was carried out based on a factorial design as described by Box et al. (1978). A high sensitivity is one requirement for an identifiable parameter the parameters with the largest impact on the model output were selected for identifiability analysis and subsequent parameter estimation.

2.2.2. Collinearity as measure for non-identifiability of parameters
The second requirement for an identifiable parameter is that a shift in the model output caused by a shift in the parameter may not be approximately compensated by appropriate changes in other parameters. The "compensability" was quantified by the calculation of the collinearity index γ_k defined by Brun et al. (2001). The collinearity index was calculated from the scaled sensitivity matrix $\widetilde{S}$.

$$\gamma_k = \frac{1}{\min_{\|\beta\|=1} \left\| \tilde{S}\beta \right\|} \quad (1)$$

The local sensitivity functions s_j were calculated for default parameter values taken from Hao et al. (2002).

2.3. Design of experiments

Different experimental layouts, operation conditions and measurements were tested and evaluated regarding identifiability of the kinetic parameters. The aim was to determine the experiment providing the most information on the selected parameters. This experiment should then be used to estimate the selected and identifiable parameters. On the one hand, continuous reactor operation (almost in steady state) was studied and compared to the results obtained for a batch experiment. On the other hand, six different experimental layouts for a batch experiment were evaluated.

3. Results and discussion

3.1. Parameters selected for identifiability analysis and parameter estimation

Regional steady state sensitivities were calculated to determine the parameters with the largest impact on the model output. The growth and inactivation rates for all three groups of microorganisms had a very large influence on the process (data not shown). All three affinity constants for oxygen had a distinct effect as well. The affinity constants for nitrite had the smallest impact on the output and were therefore not selected for parameter estimation. In total, nine parameters were selected for parameter estimation: the growth and inactivation rates for all three groups of microorganisms and all three affinity constants for oxygen.

3.2. Parameter identifiability: Continuous reactor operation versus batch experiment

Parameter identifiability was studied for continuous reactor operation and a batch experiment carried out for a dissolved oxygen concentration (DO) of 0.7 mg O_2/L and an ammonium influent and initial concentration, respectively, of 150 mg NH_4-N/L. Collinearity indices were calculated using Eq. (1) for all possible combinations of ammonium (NH_4^+), nitrite (NO_2^-) and nitrate (NO_3^-) measurements. Parameter subsets with a collinearity index below a threshold of 15 were considered identifiable.

From continuous reactor operation parameter subsets with a maximum of four parameters were identifiable from NH_4^+, NO_2^- or NO_3^- measurements (Figure 1 a). Combining measurements of the different chemical compounds did not enhance parameter identifiability as parameter subsets with a maximum of only three parameters were identifiable from combined measurements (Figure 1 b). The "good" information obtained from NH_4^+ measurements were compensated to a certain extent by the "poor" information derived from NO_2^- and NO_3^- measurements. From the batch experiment parameter subsets of size 6 could be identified from a combination of all measurements (NH4/NO2/NO3) (Figure 1 c and d). In this case, combining measurements of the different chemical compounds improved the parameter identifiability. The nine selected parameters were, however, not uniquely identifiable based on measurements from continuous reactor operation or from the batch experiment. Even combining the measurements from batch and continuous experiments did not enhance parameter identifiability. The "poor" information derived from continuous experiments compensated to a certain degree the "good" information obtained from batch experiments.

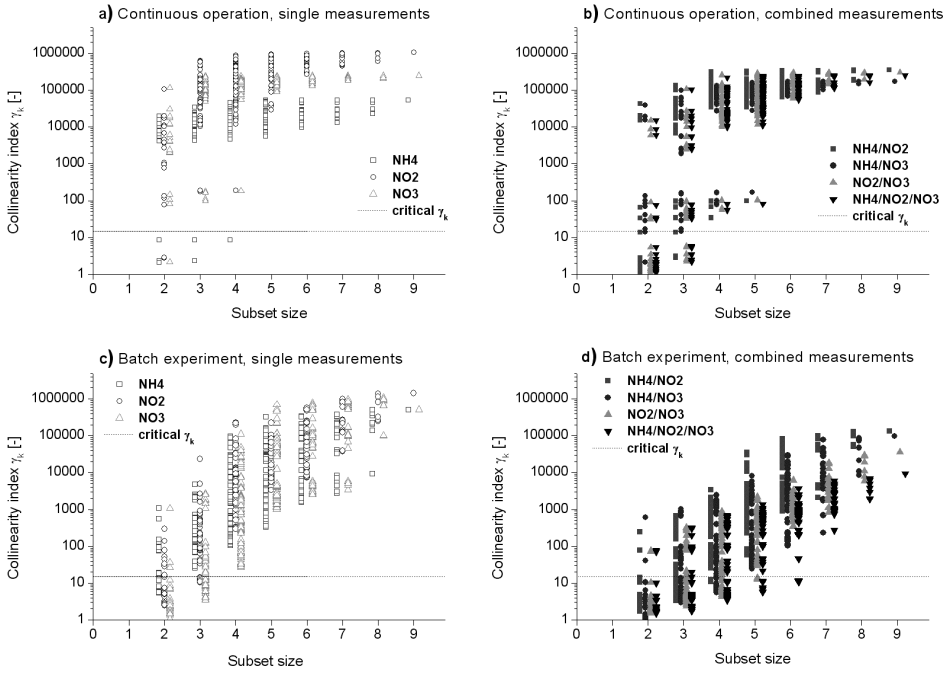

Figure 1. Collinearity indices for all parameter subsets for continuous reactor operation and batch experiment at DO 0.7 mg/L

3.3. Experimental design for batch experiments

For the experimental design six different batch experiments were investigated regarding parameter identifiability (Table 1). Besides three different dissolved oxygen concentrations in the bulk liquid (No. 1-3), the addition of nitrite at the start of the experiment (No. 4), the pulse injection of ammonium during the experiment (No. 5) and switching off the aeration at the halftime of the experiment (No. 6) were analysed concerning identifiability.

Table 1 summarises the number of identifiable subsets of parameters for NO_3^- measurements and all possible measurement combinations. These measurements and measurement combinations provide significantly more information for the identifiability analysis and the estimation of parameters compared to NH_4^+ and NO_2^- measurements. The numbers of identifiable parameter subsets are given for parameter subsets of size 5 or 6 parameters. Except for experiment No. 4, a maximum of six parameters was identifiable from the studied experimental designs based on a combination of all three measurements. Although variations of the experimental design enhanced the identifiability of the parameters for some measurement combinations not all nine selected parameters could be identified from the experimental data for any of the experimental designs. For each group of microorganisms high collinearity was observed between the growth rate and the affinity constant for oxygen. Due to the correlation between the parameters of these three subsets of size 2 only six of the nine selected parameters were identifiable.

Table 1. Number of identifiable parameter subsets of size 5 (left) or 6 (right) parameters for the investigated experimental designs

No.	Experimental design	NO3	NH4/NO2	NH4/NO3	NO2/NO3	NH4/NO2/NO3
1	DO 0.7	/ ; /	/ ; /	/ ; /	16 ; /	38 ; 8
2	DO 2	/ ; /	/ ; /	/ ; /	/ ; /	36 ; 8
3	DO 5	/ ; /	/ ; /	/ ; /	/ ; /	36 ; 8
4	DO 0.7, NH4 + NO2	/ ; /	/ ; /	/ ; /	/ ; /	36 ; /
5	DO 2, NH4 pulse	43 ; 10	10 ; /	6 ; /	24 ; /	36 ; 8
6	DO 5 off after 3 h	8 ; /	/ ; /	/ ; /	8 ; /	36 ; 8

4. Conclusions

Batch experiments provide significantly more information for the identifiability analysis and the estimation of parameters compared to continuous reactor operation at steady state. Careful selection of measurements or measurement combinations as well as the experimental design improves the identifiability of the selected parameters. Nevertheless, due to high correlations among some of the selected parameters not all of them may be identifiable from the data. To solve the parameter identifiability problems either more parameter values have to be assumed based on values from literature or parameter combinations need to be defined for highly correlated parameters.

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