IDENTIFIABILITY AND IDENTIFICATION OF A WATER TREATMENT DENITRIFYING BIOFILTER MODEL

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Abstract. This paper is concerned with the identification of the parameters of the distributed parameter model of a fixed bed denitrifying biofilter. It is twofold : first, the analysis of the structural identifiability in order to emphasize which parameters are a priori identifiable; then the identification of the model parameters by using experimental data.

Keywords. Modeling; structural identifiability; identification; water pollution

1. INTRODUCTION

Denitrification plays an important role in the treatment of drinkable water, and is largely motivated by the possibly high concentrations of nitrogen in drinkable water which may be particularly detrimental to human health. Indeed the quality of the presently available drinkable water is largely determined by the damaging effects of human activities, e.g. the intensive use of nitrogen based fertilizers not only in agriculture but also for domestic use. The regulations about the nitrogen content of drinkable water and their control tend to be more and more stringent.

Fixed bed biofilters are destined to play an increasing role in the denitrification of drinkable water. The main advantages of the fixed bed biofilters are the compactness of the process, their efficiency for biological water treatment, their good integration in the environment, and their low energy consumption.

The advent of more stringent criteria on nitrate and nitrite contents of distributed water along with the need to control and optimize the addition of a carbon source (needed to denitrify), make the use of dynamic models able to represent the behaviour of the process not only of interest but compulsory for control. If many models for the description of the biofilm growth have been proposed, very few identified models describing the biofiltration process have been published in the literature [Lessard and Beck, 1995]. If the dynamical model is considered for control design and/or for process simulation, it has to be reliable and able to reproduce as best as possible the process dynamics for the largest set of operating conditions possible. The dynamical model considered is basically derived from mass balance considerations. The biochemical reaction kinetics are assumed to be of the Monod-type, in accordance with the experiments. This results in a nonlinear distributed parameter model presented in Section 2.

The validation of the process model requires the identification of the model parameters. Because the inherent complexity of the model, an essential step and challenging question before carrying out the identification is to check whether the model is structurally identifiable, i.e. if it is theoretically possible to give unique values to the model parameters : this will be the object of Section 3. Section 4 is then dedicated to the identification of the nonlinear model parameters by using a Marquardt optimization routine.

2. DYNAMICAL MODEL OF THE BIOFILTER

2.1 *Description of the biofilter*

The considered denitrification process is a submerged granular fixed-bed biofilter of 2.1m high (Fig. 1).

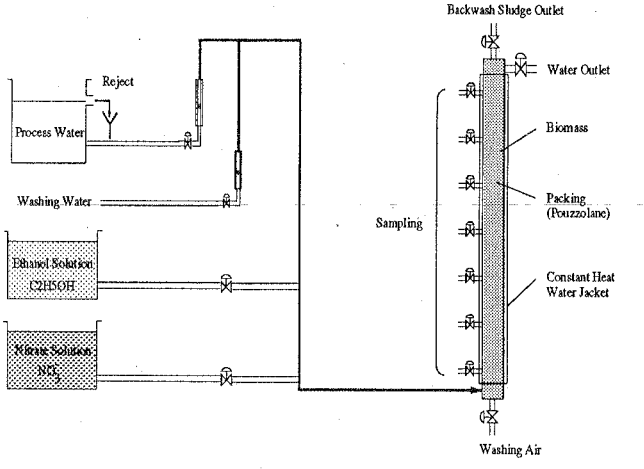

Fig. 1. *Schematic view of the denitrification process*

It is packed with pouzzolane, a very porous volcanic material which retains a big amount of microorganisms during the washing. Thus the initial biomass concentration is often very close to the maximum active biomass concentration. The substrates to be removed can freely move along the reactor. The inside temperature is almost constant due to a double insulation enveloppe. A solution of ethanol (needed for the biological reactions) is mixed with the feeding nitrate in a tank at the inlet of the reactor. Eight measurement points are distributed at every thirty centimeters along the column for the measurement of nitrate and nitrite concentrations. In some experiments, off-line measurements of ethanol are also performed from samples at the eight measurement points. Preliminary experiments in open loop showed that the axial dispersion phenomenon is negligible, and that the bioreactor is basically operating in plug flow conditions.

2.2 *The dynamical model*

The denitrification processing is defined as being the reduction of nitrates (electron acceptor) NO_3^- into gaseous nitrogen N_2 by using organic carbon (electron donor) with production of intermediate compounds, namely nitrites NO_2^- ($NO3^- \xrightarrow{(1)} NO2^- \xrightarrow{(2)} N_2$). In order to dissociate nitrates and nitrites, two distinct biological reactions are considered in the model, denitratation and denitritation.

The denitrification processing is a reaction which is performed in anaerobic conditions (medium without air nor oxygen). The gaseous phase is not taken into account because the gaseous hold-up may be neglected compared to the liquid one. A mass balance dynamical model taking into account biological reaction schemes and deep filtration has been developped and published in ([JACOB, 1994]) :

$$\frac{\partial S_1(z,t)}{\partial t} = -\frac{u}{\varepsilon}\frac{\partial S_1(z,t)}{\partial z} - \frac{k_1 - 1}{\varepsilon\alpha_1}\mu_1 X_a(z,t) \quad (1)$$

$$\frac{\partial S_2(z,t)}{\partial t} = -\frac{u}{\varepsilon}\frac{\partial S_2(z,t)}{\partial z} + \frac{k_1 - 1}{\varepsilon\alpha_1}\mu_1 X_a(z,t) - \frac{k_2 - 1}{\varepsilon\alpha_2}\mu_2 X_a(z,t) \quad (2)$$

$$\frac{\partial S_3(z,t)}{\partial t} = -\frac{u}{\varepsilon}\frac{\partial S_3(z,t)}{\partial z} X_a(z,t) - \frac{1}{\varepsilon}\left(k_1\mu_1 + k_2\mu_2\right) X_a(z,t) \quad (3)$$

$$\frac{\partial X_a(z,t)}{\partial t} = [\mu_1 + \mu_2]\left(1 - \frac{X_a(z,t)}{X_{a,max}}\right) X_a(z,t)$$

Boundary conditions are given at the input of the process : $S_l(z = 0, t) = S_{l,in}(t)$ for $l = 1, 2, 3$ and $X_a(z = 0, t) = 0$.

In the above equations, ε, $S_1(z,t)$, $S_2(z,t)$, $S_3(z,t)$, $X_a(z,t)$, $X_{a,max}$, u, and μ_i represent respectively the porosity, concentrations of nitrate, nitrite, carbon, active biomass, maximum biomass (g/m^3), the fluid superficial velocity (m/h), and the specific growth rates ($g/m^3/h$). The specific growth rates are described with the double Monod-type model :

$$\mu_i = \mu_{i,max}\frac{S_i}{S_i + K_{mi}}\frac{S_3}{S_3 + K_{m3}} \quad \text{for} \quad i = 1, 2 \quad (4)$$

where $\mu_{1,max}$ and $\mu_{2,max}$ are the maximum values of the specific growth rates μ_1 and μ_2 ; K_{m1}, K_{m2} and K_{m3} are the Monod constants associated to nitrate, nitrite and carbon respectively. Coefficients k_i are defined as follows : $k_1 = \frac{1}{Y_{h1}}$, $k_2 = \frac{1}{Y_{h2}}$.

2.3 *Model for the identifiability analysis and identification*

The real denitrification process is normally running around a steady state or subject to conditions where

the time variations in the system are relatively slow and small. Moreover the measurements of the different concentrations of nitrate, nitrite and carbon are performed at the end of each operation cycle, i.e. when the active biomass concentration X_a has reached its maximum value $X_{a,max}$ ($X_a = X_{a,max}$). Then a feasible approximation from the identification point of view is to study the system in the state where the partial derivatives of the state variables with respect to the time are set equal to zero :

$$-u\frac{\partial S_1(z,t)}{\partial z} - \frac{k_1 - 1}{\alpha_1}\mu_1 X_a(z,t) = 0 \quad (5)$$

$$-u\frac{\partial S_2(z,t)}{\partial z} + \left[\frac{k_1 - 1}{\alpha_2}\mu_1 - \frac{k_2 - 1}{\alpha_2}\mu_2\right] X_a(z,t) = 0 \quad (6)$$

$$-u\frac{\partial S_3(z,t)}{\partial z} - [k_1\mu_1 + k_2\mu_2] X_a(z,t) = 0 \quad (7)$$

In the above model (5)(6)(7)(4), there are eight parameters : $\mu_{i,max|i=1,2}$, $K_{mi|i=1,2,3}$, $k_{i|i=1,2}$ and $X_{a,max}$. Finally two types of experimental data sets are available: one with the three substrates S_1, S_2 and S_3, the other where only measurements of S_1 and S_2 have been performed.

3. STRUCTURAL IDENTIFIABILITY

3.1 *Introduction*

The notion of structural identifiability is related to the possibility to give a unique value to each parameter of a mathematical model. In simple words, the structural identifiability of a model can be formulated as follows ([GODFREY and DISTEFANO, 1987]) : given a model structure and perfect (i.e. that fit perfectly to the model) data of model variables, are all the parameters of the model identifiable? From the structural identifiability analysis one may conclude that only combinations of the model parameters are identifiable. If the number of resulting combinations is lower than the original model parameters, or if there is not a one-to-one relationship between both parameter sets, then a priori knowledge about some parameters may be required to achieve identifiability.

For linear systems, the structural identifiability is rather well understood, and besides classical identifiable models (like dynamical models in canonical form), there exists a number of tests for the identifiability (e.g. Laplace transform method, Taylor series expansion of the observations, Markov parameter matrix approach, modal matrix approach, ... [GODFREY and DISTEFANO, 1987]). However, for models that are nonlinear in the parameters (like the models used in this work), the problem is much more complex. There exist also several structural identifiability tests, but they are usually very complex (they typically require the (very helpful) use of symbolic software packages, as will be illustrated below). In the following several similar approaches are used wherein the models are transformed into linear ones, after which the analysis is based on the linear model.

Here we have considered three different approaches for testing the structural identifiability of (5)-(7), (4).

- The Taylor series expansion consists of expanding in Taylor series the output function $y(z)$ around $z = 0$ [GODFREY and DISTEFANO, 1987] [POHJANPALO, 1978], and then of calculating successive derivatives of this function, to check if they contain information about the parameters (or parameter combinations) to be identified.

- Another approach for analyzing the structural identifiability is to transform the nonlinear model into a model linear in the parameters, and then look at the identifiability of the linear model ([DOCHAIN et al., 1995]).

- The third considered approach is based on the theory of the observability of nonlinear systems : indeed the study of the identifiability properties of a nonlinear system can be reduced to the study of the observability properties of the nonlinear system augmented by an auxiliary vector containing the model parameters to be identified ([WALTER and PRONZATO, 1995]).

The approaches have been considered for both types of experiments, i.e. once when the output y_1 is equal to :

$$y_1 = [S_1,\ S_2,\ S_3]^T \quad (8)$$

and once when the output contains two substrates :

$$y_2 = [S_1,\ S_2]^T \quad (9)$$

And in both cases, the Maple symbolic calculation software has been used for the successive symbolic calculations. A careful look at the structure of the model equations shows that the parameters $X_{a,max}$ and $\mu_{1,max}$, and $X_{a,max}$ and $\mu_{2,max}$ always intervene under the form of a product : this implies that these parameters cannot identified separately. For the first case, we have concluded that the following set of seven parameter combinations is structurally identifiable :

$$\vartheta_1 = \mu_{1,max}X_{a,max}, \vartheta_2 = \mu_{2,max}X_{a,max},\ \vartheta_3 = k_1,\ \vartheta_4 = k_2,\ \vartheta_5 = K_{m1}, \vartheta_6 = K_{m2},\ \vartheta_7 = K_{m3}$$

For the second case, we have not been able to conclude. However we knew from a preliminary sensitivity analysis showed that the output of the model is relatively insensitive to the changes in the saturation constants K_{mi} of the Monod model. Therefore we decided to restart the analysis without them. The conclusions with the three approaches is then that the following parameter combinations are structurally identifiable :

$$\vartheta_1 = \mu_{1,max} X_{a,max}, \vartheta_2 = \mu_{2,max} X_{a,max}, \ \vartheta_3 = k_1, \ \vartheta_4 = k_2,$$

The detailed calculations of the identifiablity analysis are presented in [BOURREL, 1996]. Here, only one case is presented, the second approach for the first output y_1.

3.2 *Transformation of the nonlinear model into a linear one in the parameters*

Let us first consider in the following the more compact notation : $\mu_{1,max} \equiv \mu_1$, $\mu_{2,max} \equiv \mu_2$, $X_{a,max} \equiv X$. The system (5)-(7), (4) may then be rewritten under the following equivalent form :

$$\frac{dS_1}{dz} = -\frac{1}{\alpha_1 u}\frac{1-Y_{h_1}}{Y_{h_1}}\mu_1 X \frac{S_1}{S_1+K_{NO_3}}\frac{S_3}{S_3+K_C} \quad (10)$$

$$\frac{d(S_1+S_2)}{dz} = -\frac{1}{\alpha_2 u}\frac{1-Y_{h_2}}{Y_{h_2}}\mu_2 X \frac{S_2}{S_2+K_{NO_2}}\frac{S_3}{S_3+K_C} \quad (11)$$

$$\frac{dS_3}{dz} = -\frac{1}{u}\frac{1}{Y_{h_1}}\mu_1 X \frac{S_1}{S_1+K_{NO_3}}\frac{S_3}{S_3+K_C} - \frac{1}{u}\frac{1}{Y_{h_2}}\mu_2 X \frac{S_2}{S_2+K_{NO_2}}\frac{S_3}{S_3+K_C} \quad (12)$$

The adavantage of the above formulation for the identifiability analysis is that each equation contains a ratio of polynomials function of S_1, S_2 and S_3 which are different for each equation. Moreover it avoids some redundancy (which appears with the original formulation (5)-(7)) in the parameter sets between the first and the second equation.

Let us multiply each equations of the system (10)(11)(12) respectively by :

- $(S_1+K_{NO_3})(S_3+K_C)$
- $(S_2+K_{NO_2})(S_3+K_C)$
- $(S_1+K_{NO_3})(S_2+K_{NO_2})(S_3+K_C)$

Then the system (10)(11)(12) becomes :

$$(S_1+K_{NO_3})(S_3+K_C)\frac{dS_1}{dz} = -\frac{1-Y_{h_1}}{\alpha_1 u Y_{h_1}}\mu_1 X S_1 S_3$$

$$(S_2+K_{NO_2})(S_3+K_C)\frac{d(S_1+S_2)}{dz} = -\frac{1-Y_{h_2}}{\alpha_2 u Y_{h_2}}\mu_2 X S_2 S_3$$

$$(S_1+K_{NO_3})(S_2+K_{NO_2})(S_3+K_C)\frac{dS_3}{dz} = -\frac{1}{uY_{h_1}}\mu_1 X S_1 S_3 (S_2+K_{NO_2}) - \frac{1}{uY_{h_2}}\mu_2 X S_2 S_3 (S_1+K_{NO_3})$$

The system can be rewritten as follows by expanding the different terms :

$$S_1 S_3 \frac{dS_1}{dz} + \beta_1 S_1 \frac{dS_1}{dz} + \beta_2 S_3 \frac{dS_1}{dz} + \beta_1\beta_2 \frac{dS_1}{dz} = -\frac{1}{\alpha_1 u}\beta_3 S_1 S_3 \quad (13)$$

$$S_2 S_3 \left(\frac{dS_1}{dz}+\frac{dS_2}{dz}\right) + \beta_1 S_2 \left(\frac{dS_1}{dz}+\frac{dS_2}{dz}\right) + \beta_4 S_3 \left(\frac{dS_1}{dz}+\frac{dS_2}{dz}\right) + \beta_1\beta_4\left(\frac{dS_1}{dz}+\frac{dS_2}{dz}\right) = -\frac{1}{\alpha_1 u}\beta_5 S_2 S_3 \quad (14)$$

$$S_1 S_2 S_3 \frac{dS_3}{dz} + \beta_1\beta_4 S_1 \frac{dS_3}{dz} + \beta_1\beta_2 S_2 \frac{dS_3}{dz} + \beta_2\beta_4 S_3 \frac{dS_3}{dz} + \beta_1 S_1 S_2 \frac{dS_3}{dz} + \beta_2 S_2 S_3 \frac{dS_3}{dz} + \beta_4 S_1 S_3 \frac{dS_3}{dz} + \beta_1\beta_2\beta_4 \frac{dS_3}{dz} = -\frac{1}{u}(\beta_6+\beta_7) S_1 S_2 S_3 - \frac{1}{u}\beta_4\beta_6 S_1 S_3 - \frac{1}{u}\beta_2\beta_7 S_2 S_3 \quad (15)$$

with : $\beta_1 = K_C$, $\beta_2 = K_{NO_3}$, $\beta_3 = \frac{1-Y_{h_1}}{Y_{h_1}}\mu_1 X$, $\beta_4 = K_{NO_2}$, $\beta_5 = \frac{1-Y_{h_2}}{Y_{h_2}}\mu_2 X$, $\beta_6 = \frac{1}{Y_{h_1}}\mu_1 X$, $\beta_7 = \frac{1}{Y_{h_2}}\mu_2 X$.

The system (13)(14)(15) is a system linear in the parameters $\beta_{i_{i=1,2,\ldots,7}}$ and depends on the values of the concentrations in nitrate, nitrite and carbon at the different positions along the denitrification column. Since the regressors (i.e. the terms multiplying the parameters β_i) are formally independent (i.e. it is formally possible to generate space dependent values of the variables S_1, S_2, S_3 and of their space derivatives such that the regressors are independent), the structural identifiability of the parameters β_i of the system (13)(14)(15) follows.

Let us write the expressions of the initial model parameters in terms of the parameters $\beta_{i_{|i=1,2,\ldots,7}}$:

$K_{NO_3} = \beta_2$, $K_{NO_2} = \beta_4$, $K_C = \beta_1$, $\mu_1 X = \beta_6 - \beta_3$, $\mu_2 X = \beta_7 - \beta_5$, $Y_{h_1} = \frac{\beta_6-\beta_3}{\beta_6}$, $Y_{h_2} = \frac{\beta_7-\beta_5}{\beta_7}$.

We can then conclude that the above seven parameters (K_{NO_3}, K_{NO_2}, K_C, $\mu_1 X$, $\mu_2 X$, Y_{h_1}, Y_{h_2}) are structurally identifiable. Note in particular, as it has been suggested hereabove, that only the products $\mu_1 X$ and $\mu_2 X$ are identifiable.

4. IDENTIFICATION

4.1 *Methodology and Experimental Data*

The identification method used is based on the minimization of a quadratic performance index by using the Marquardt method [MARQUARDT, 1963] modified for the dynamic case ([NIHTILÄ and VIRKKUNEN, 1977]) ; the knowledge of the sensitivity functions with respect to the parameters $\vartheta_{i|i=1,\ldots,4}$ is needed. The performance index is a quadratic function of the error between calculated and measured concentrations of nitrate and nitrite at some internal points of the reactor and at the outlet.

As already mentioned in the previous section, a preliminary sensitivity analysis showed that the output of the model is relatively insensitive to the changes in the saturation constants K_{mi} of the Monod model. Consequently, we decided to fix the values of these parameters to constant values (found in the literature). This leaves four parameters to be identified :

$$\mu_{i,max|i=1,2} X_{a,max} \quad \text{and} \quad k_{i|i=1,2}$$

The integration of the differential system was done by using a Runge-Kutta method. An identification software package written in C provides values to unknown parameters $\vartheta_{i|i=1,\ldots,4}$.

Real data on concentrations were obtained from 14 systematically planned experiments performed on the pilot-scale denitrification process. In these experiments, the concentration in nitrogen in the inlet was equal to 16.93 $g[N]m^{-3}$ ($S_{1,in} = 16.93\ g[N]m^{-3}$, $S_{2,in} = 0$). The value of the inlet ethanol concentration has been chosen so as to have a ratio of carbon over nitrogen (C/N) equal to 1.5. The parameter identification has been performed with the following numerical values for the affinity constants :

$$K_{m1} = 2\ g.m^{-3} \quad K_{m2} = 3\ g.m^{-3} \quad K_{m3} = 40\ g.m^{-3}$$

The identification proceeds in two steps. First of all, the calibration of the model parameters is performed on one set of the experimental data by using the Marquardt algorithm. Then the estimated values of the parameters are validated on the remaining set of experimental data (by comparing the experimental data with numerical simulation of the model with the estimated parameter values).

For the fitting of the estimation curves on the experimental data, we have taken into account the value of the measurement errors on the concentrations in nitrates, nitrites and ethanol; this error has been estimated to be equal to $\varepsilon_{\text{mes}} = \pm\ 0.5\ g\ N/m^3$ for the concentrations in nitrate and in nitrite and to $\varepsilon_{\text{mes}} = \pm\ 2\ \%$ (of each measurement) for the ethanol concentrations.

Moreover let us define :

• the error at each measurement points (z_{mes_j}) at the final time t_f (before washing) between the estimation (s_i for i=1,2,3) and the experimental data at these points,

$$\varepsilon_i(z_j, t_f) = s_{i_{mes}}(z_{mes_j}, t_f) - s_i(z_{mes_j}, t_f) \quad \text{for i = 1,2,3}$$

• the mean error (ε_{imoy}), the variance (Var_i) and the standard deviation (σ_i) with p_{mes} the number of measurement points :

$$\varepsilon_{imoy} = \frac{1}{p_{mes}} \sum_{j=1}^{p_{mes}} s_{i_{mes}}(z_{mes_j}, t_f) - s_i(z_{mes_j}, t_f)$$

$$\text{Var}_i = \frac{1}{p_{mes}} \sum_{j=1}^{p_{mes}} (\varepsilon_i(z_{mes_j}, t_f) - \varepsilon_{imoy})^2$$

$$\sigma_i = \sqrt{\text{Var}_i} \quad \text{for i = 1,2,3}$$

Finally the identification algorithm has been implemented so as to allow the identification of the inital conditions $S_1(z=0)$, $S_2(z=0)$ and $S_3(z=0)$. Indeed it has been experimentally noted that there is a consumption of organic carbon by the oxygen present at the bottom of the column. Since this consumption is not considered in the model, this might lead to discrepancies in the identification and therefore we decided to allow to complement the parameter identification with that of the initial conditions. In the following curves, the dotted lines represent the curves simulated from the model parameter identification with taking into account the initial conditions; the straight lines account for the initial conditions. "+" and "o" represent the errors at each measurement point with and without the identification of the initial conditions, respectively.

4.2 *Identification results*

□ **Identification :** The following table summarizes the values obtained by the identification with and without the initial conditions being identified for one experiment (13/04/95). The values of the estimated parameters are in line with those generally considered and admitted in the scientific literature. In the following, "i.c." means initial conditions (when initial conditions are identified).

	Algorithm initialisation	After identification no i.c.	i.c.
$S_{NO_3}(z=0)$	16.93	- -	17.23
$S_C(z=0)$	101.56	- -	98.00
$\mu_{NO_{3max}} X_{a,max}$	250	318.23	269.85
$\mu_{NO_{2max}} X_{a,max}$	250	168.01	159.75
Y_{h_1}	0.6	0.6	0.55
Y_{h_2}	0.6	0.42	0.40

Table 1 : Results of the parameter identification for one experiment (13/04/95)

Note that in the present case, the fitting of the model on the data with and without the initial conditions are close to each other (see Table 2 and Fig.2), and that the measurements of the initial conditions are yet quite close to the estimated values. Table 2 summarizes for the same experiment the values of ε_{imoy} and σ_i for the concentrations in nitrate, nitrite and ethanol:

	Experiment of 13/04/95					
	ε_{1moy}	σ_1	ε_{2moy}	σ_2	ε_{3moy}	σ_3
no i.c.	0.3587	0.3476	-0.1563	0.5122	-1.6402	5.6045
i.c.	0.2916	0.3204	-0.1574	0.4649	-1.7844	4.9390

Table 2 : Mean values and standard deviations on the profiles of nitrates and nitrites for the experiment (13/04/95)

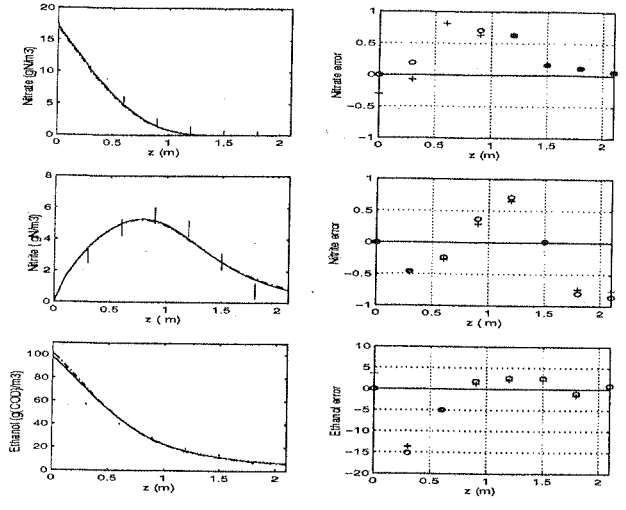

Fig. 2. *Identification results for one experiment (13/04/95)*

□ **Validation :** The validation of the estimated parameters are validated on the remaining set of experimental data (see Figure (3)). Figure (3) shows that for these parameters, the deviations between the simulated curves and the experimental profiles are very low. Table 3 summarizes for the same experiment the values of ε_{imoy} and σ_i for the concentrations in nitrate, nitrite and ethanol.

	Experiment of 05/04/95					
	ε_{1moy}	σ_1	ε_{2moy}	σ_2	ε_{3moy}	σ_3
no I.C.	0.3550	0.6352	0.0174	0.6778	-5.1502	6.6226
I.C.	0.2879	0.5499	0.0164	0.6960	-5.2944	6.0174

Table 3 : Mean values and standard deviations on the profiles of nitrates and nitrites for the experiment (05/04/95)

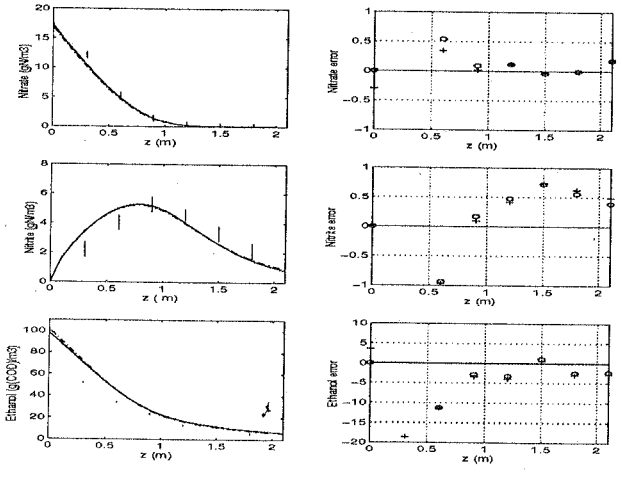

Fig. 3. *Validation on the experiment (05/04/95)*

5. CONCLUSIONS

This paper was concerned with the identification of the parameters of the distributed parameter model of the fixed bed biofilter. It was twofold : first, the analysis of the structural identifiability in order to know which parameters are a priori identifiable; then the identification of the model parameters by using experimental data. This study has been used to design and implement an adaptive linearizing controller ([BOURREL, 1996]).

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