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FROM DATA TO MECHANISTIC-BASED MODELS

OD WYNIKÓW EKSPERYMENTU DO MODELU MECHANISTYCZNEGO

Abstract

In this work, an Approach to Mechanistic Identification is proposed which facilitates an efficient simulation of complex dynamic processes. The developed algorithm divides the process in multiple intervals each of which is simulated by a different model in order to overcome the limitations of every single model and to take advantage of its capacities. Furthermore, the approach applies mechanistic models. By this means it enables a better understanding of the process subject to study by clarifying the predominant phenomenon inducing the process behaviour in each considered interval. The problem is set as an optimization problem whose objective function is a least square error function. The parameters of the model and the size of the intervals are estimated to find the "best" fit.

Keywords: complex dynamic processes, simulation, mechanistic identification, predominant phenomenon

Streszczenie

W niniejszym artykule zaproponowano założenia mechanistycznej identyfikacji dla skutecznej symulacji złożonych, dynamicznych procesów. W opracowanym algorytmie proces podzielono na przedziały czasowe, w których przebiegające procesy opisane są stosownymi, różnymi, modelami. Zastosowanie mechanistycznego podejścia umożliwia lepsze zrozumienie procesu i określenie dominującego zjawiska określającego jego przebieg. Problem zastosowano w optymalizacji, gdzie parametrem jest błąd funkcji określany metodą najmniejszych kwadratów. Parametry modelu i długość przedziałów czasowych szacowane są dla zapewnienia najlepszego dopasowania.

Słowa kluczowe: złożone procesy dynamiczne, symulacja, identyfikacja mechanistyczna, dominujące zjawisko

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1. Introduction

A novel framework for mechanism identification has been designed as a response to the actual exigencies of process engineering. The chemical industry in our days faces its most competitive era. Globalization, environment restrictions and the uncertainty in the prices of energy sources permit only the most efficient processes to be profitable. This has led to an exponential grow in the complexity of processes applied in the industry. In addition, the quality of control engineering, plant design and research and development of a process depends directly on the model used to describe it. This combination commits model engineers with the epic task to provide precise simulation of complex dynamic systems. Two fields are especially affected by this problem, Catalysis and Biotechnology.

Catalysts present in our days the most promising short-term solution to reduce the energy consumption of processes as well as the pollution they produce [1]. Chemical reactions are difficult to simulate, not only because of the many physicochemical phenomena taking part in the process, e.g. adsorption, but also because of the dynamical behaviour of catalyst fouling. An example of this is the most successful process for gasoline production Fluid Catalytic Cracking (FCC). This process has been used to obtain gasoline from heavy hydrocarbons since 1936. Although restless research has been made in this topic, no definitive model capable of simulating a dynamic FCC process is available in the literature [2].

Advances in biotechnology confront simulation with a new and extensive branch of problems to be solved by engineers. Biological processes have an immense number of reactions, most of them unknown, which depend on factors that are not completely understood. In E. coli batch fermentations, for example, the production of acetate depends on the amount of glucose and the growth rate among many other parameters. The fermentation presents a production of acetate under glucose excess and a consumption of acetate under lack of nutrients [3]. The reason for E. coli to excrete acetate is still not well understood and novel procedures to enable the monitoring of the acetate production are still under investigation [4]. Nonetheless, E. coli fermentation for recombinant production of proteins is the standard process for Humulin, synthetic insulin [5]. Therefore, a model with the capability to simulate the production of acetate in such fermentations is not yet available and extremely useful.

Many models have been developed to simulate processes with these characteristics. Nevertheless, the results obtained until our days present either an extensive and with high calculation costs algorithm or imprecise outputs. The basic problem which obstructs a simple and optimal simulation is the fact that many different phenomena take role in these processes and induce a dynamical behaviour. Most processes can be effectively simulated only for short periods of time but not for the whole process.

In order to overcome these problems, a first attempt towards mechanism identification has been proposed in [6]. The approach considers many different models and uses them in such a way that each model is applied in the precise moment in which it best describes the process. A similar concept was published by Keneth D. Forbus in 1981 [7]. In this paper Forbus proposes the Qualitative Process theory (QP), a model discrimination based on qualitative information. Disadvantages of this method is that no quantitative analysis is realized, most of the processes can not be described with such simple concepts as positive--negative slope or open-closed valve. Neither semi-quantitative programming can be

applied in most processes. Quantitative variables or changing parameters can frequently not be measured neither directly nor indirectly with the precision needed e.g. catalyst fouling or bacterial stress.

The proposed approach in this contribution substitutes the qualitative concept of QP with a quantitative Model Discrimination as used in the Optimal Experiment Design (OED) to select the appropriate model. OED is a statistical method used to reduce the costs of experimental work needed to select the model which best describes the investigated topic. An analysis of the correlations and variances of the parameters of the model determines how many experiments have to be realized and which inputs and initial conditions provide the most information [8]. These mathematical tools can be used to select the best model among many in a specific interval.

Allying the methods used in common parameter optimization, the well known and widely applied Optimal Experiment Design (OED) and the basic approach proposed by the QP theory, a new concept arises and presents a novel solution to actual problematic in the simulation of complex engineering processes.

2. Solution approach

Based on the developed approach, the suitable model to each process interval is to be found aiming to describe the integral process in an optimal way. In order to obtain the perfect model combination as well as the exact number and time length of each interval, an exhaustive analysis of every point in a long iterative process is necessary. It is clear that a conciliation point between accuracy and calculation time has to be found.

The method can basically be divided in three main steps:

– interval selection,
– model discrimination,
– optimization of the interval limits.

Interval selection

The first necessary step is to make a selection of the number of interval in which the process will be divided as well as the approximated time range of each interval. This decision is made based on an analysis of the experimental data available. The number of models proposed, the behaviour of the data and the experience of engineer are the tools available for these decisions to be made.

Model discrimination

Mechanistic models which are considered to be capable of describing the process are called candidate models. Candidate models are set to discrimination until one model is selected, all models are rejected or it is considered that the information is not enough to make a model selection. The discrimination of the models follows the OED method. The models are first set to a parameter analysis to determine:

Identifiability: whether every change on the parameters is reflected in the model behaviour.

Distinguishability: the capability of every model to show a different response at the same initial conditions and inputs.

When one model is selected the next step is followed. If no model is selected or more than one show to be adequate further experimentation data is needed or the time intervals have to be extended.

For parameter estimation an optimization problem with the objective function Eq. (1) is defined

$$\Phi(p_{es}) = \frac{1}{2}\sum_{i=1}^{n}\sum_{k=1}^{N}\frac{(x_i(p_{es},t_k)-z_{i,k})^2}{\sigma_{i,k}^2} + \frac{1}{2}\sum_{k=1}^{N}\ln(\det(C(t_k))) + C1 \tag{1}$$

where:

Φ – the objective function;
p_{es} – estimated parameters;
x – predicted values;
t_k – the time at measure number k
z – the measured value;
σ^2 – variance of the error at point i, k;
C – the covariance matrix.

Because the constant value has no effect on the position of the optimal parameter vector and the covariance matrix is considered as known, the function used in the algorithm is Eq. (2)

$$\Phi(p_{es}) = \frac{1}{2}\sum_{i=1}^{n}\sum_{k=1}^{N}\frac{(x_i(p_{es},t_k)-z_{i,k})^2}{\sigma_{i,k}^2} \tag{2}$$

By derivation of the objective function with respect to its parameter vector, we obtain the Fisher information matrix. The Fisher matrix enables an effective analysis of the parameters. The Fisher matrix eq. (3) shows the eigenvalues of the matrix

$$F(p_{es}) = \sum_{k=1}^{N}\left(\frac{\partial\Phi}{\partial p}\right)_{p_{es},t_k} C_M^{-1}(t_k)\left(\frac{\partial\Phi}{\partial p}\right)_{p_{es},t_k}^{T} \tag{3}$$

where:

F – Fisher Information Matrix with

$$C_M = E\left\{(z_{i,k} - x(p_{es},t_k))(z_{i,k} - x(p_{es},t_k))^T\right\} \tag{4}$$

This procedure is equivalent to the calculation of the basis vectors of a coordinate system. It has also the same objective. The fisher matrix allows seeing in a separated way the influence of every parameter in the general probability function. The smallest value in the diagonal of the matrix is also the value with the highest variance. There are multiple optimization approaches to search for the result with the smallest probability function. Commonly used are the A, D and E-criteria [9]. A graphical example can be seen in Fig. 1.

Once a model has been selected for each interval, the optimization of the interval boundaries should reduce the objective function which can be described as a least square error function. This is also the most critical step of the optimization due to its important effect in the overall optimization. It is also a discrete, stiff, nonlinear function which presents a high optimization challenge.

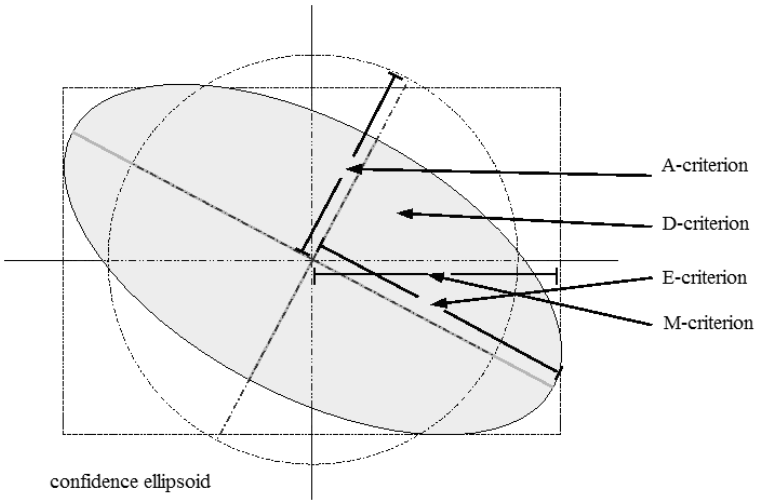

Fig. 1. Optimizing the interval lengths

Rys. 1. Optymalizacja długości przedziału czasu

3. Case study: a membrane bioreactor

The example selected to show the efficiency of the proposed approach is a bioreactor. The resulting framework was applied and it was a proficient way to successfully simulate the entire fouling process of a membrane in the Membrane Bioreactor (MBR) at constant pressure. Although the algorithm has been subject to different improvements concerning the flow diagram and the optimization tools implemented, it is still in development due to the challenges concerning their broad application to catalytic and biocatalytic processes.

3.1. Membrane bioreactor

Membrane Bioreactors (MBR), consist of a biological reactor joined to a filtration process. MBR technology has undergone a great development during the course of the last years and is being implanted to the detriment of the traditional decantation separation method.

The application of these technologies allows the separation of the active sludge from the liquid through membranes, with a method that has been proven to be robust, reliable and can provide high quality water [10].

The operation is based on the filtration of the liquid coming from the biological reactor. The permeate (purified water) crosses the membrane walls leaving the retentate (active sludge) in the reactor. The filtered water is extracted from the system while the mud and big size particles retained by the membrane are returned to the biological reactor. In Fig. 2 a scheme of a MBR is shown.

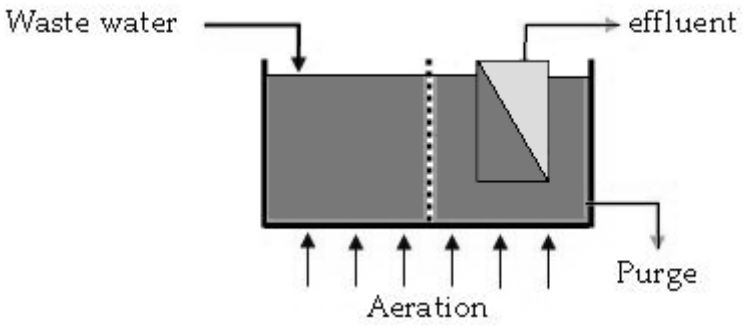

Fig. 2. MBR scheme

Rys. 2. Schemat reaktora MBR

When the membrane is dirty, three different processes are applied in order to regenerate the membrane:
– a filter pause is performed,
– a counter-flow wash is applied by changing the direction of the filtered water and detaching particles from the surface of the filter,
– a periodically regeneration that consists in submerging the membrane in solutions with detergents and sodic hypochlorite among other products.

3.2. Mechanistic filtration models

Five models are subject to discrimination in each interval. The first is also the most common model named Dead End J. Hermia [11] proposed three models called Standard Intermediate and Complete Blocking. Finally, the model called Cross-Flow developed by M.G. Liu [12] is also considered. With the help of the approach, the parameters of each model are estimated and the one which best describes each interval is then selected.

Dead End

This model is based on the hypothesis that the pressure drop is caused by:
– the pressure drop due to membrane which will be constant along the time,
– the cake resistance which may be variable due to membrane fouling, increasing along the time.

Standard Blocking Filtration Law

This model is based on two assumptions. On the one hand, the pore volume is reduced proportionally to the filtrated volume due the deposition of particles inside the membrane. On the other hand, the pores have the same values of diameter and length.

Intermediate Blocking Filtration Law

In the intermediate blocking law, the particles can block a pore becoming an impermeable surface, or can settle on deposited particles. For each possibility there is the same probability, this means that the proportion of blocked area is the same than the clean, which will decrease gradually.

Complete Blocking Filtration Law

In this model it is assumed that every particle which reaches the membrane surface blocks a pore and when a pore it is blocked it becomes impermeable. Consequently, the complete blocking filtration law reduces the active filtration area faster than other models.

Cross Flow

Several bibliographical sources [13] – refer this model which is based on the hypothesis that the particles have different behaviours along the filtration process:
– particles deposited on the membrane's surface,
– particles that contribute to the internal clogging,
– particles transported from the membrane's surface to the liquid phase in a retroflow process which cleans the membrane.

3.3. Solution strategy

The solution approach has been developed in a Matlab® environment. By this means, a framework has been implemented so as to automatically recognize the mechanisms in an activated sludge filtration processes at constant pressure. The algorithm first chooses the adequate model for each interval. Once this step is successfully completed, the program shifts the interval borders to reduce the least square error between data and model prediction.

It is considered that the filtration is started with a clean membrane through which water is filtered leaving the activated sludge in the reactor. During this filtration process the sludge is accumulated on the membrane surface creating a filtering cake. This accumulation increases the pressure loss in the membrane reducing the water flow at constant driving force.

Figure 3 shows data measured on a lab-scale (test cell system) at the laboratory. A detailed description of the experiment can be found in [14].

Using the data shown in Fig. 3, the program begins dividing the time in five equidistant intervals. In this case the number of intervals was selected considering the number of candidate models. Once the boundaries have been determined, the program estimates the parameters of each model aiming to reduce the difference between the mathematical models and the laboratory data of each interval. A Quasi Newon-Rapson algorithm is used to minimize the objective function, which is the sum of the square of errors. The generated Least Square Error (LSQ) is computed and used to select the best model. Once the model discrimination has ended and every interval has been assigned to a model, the next optimization step is the determination of the optimal position of the interval boundaries. For

this purpose, the Nelder-Mead simplex algorithm [15] is applied. This method finds the global minima by a direct function evaluation. The algorithm is very robust since no Jacobi matrix has to be calculated and can overcome local minima with a high efficiency. An example of this procedure can be seen in Fig. 4.

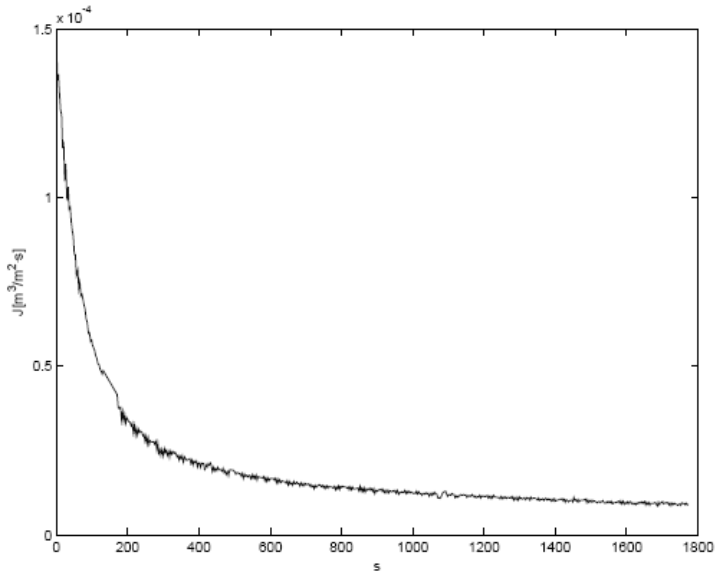

Fig. 3. An example of experimental data

Rys. 3. Przykładowe wyniki eksperymentu

The error has to be evaluated at the end of each iteration so as to check whether this error is over or under the maximum tolerance. The difference between the data and the mathematical models has to be always positive. The objective function is then given as follows

$$Error = \sum_{i=1}^{n} (B_i - C_i)^2 \tag{5}$$

where:

B – experimental data,

C – data obtained with mathematical models.

The solution strategy is illustrated in Fig. 5.

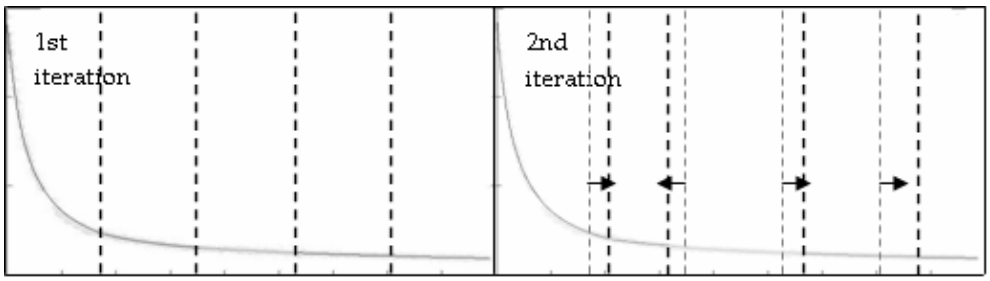

Fig. 4. Movement of time intervals

Rys. 4. Przesunięcie przedziałów czasowych

4. Results

The developed framework was implemented for the prediction of 6 experiments. The data obtained in the experiments represents the input. The Minimal Square Error was in an order of magnitude between $7{,}43e^{-10}$ as the greatest value and $3{,}81e^{-11}$ as the best achieved. The two most illustrative results are exposed as follows.

Experiment 1

Experiment 1 is the experiment with the lowest variance. The framework showed an LSQ = $6{,}30e^{-10}$ which is almost ten times better than the LSQ = $4{,}56e^{-9}$ showed by the Complete Blocking model which was the most effective. Based on the model sequence, the approach can also describe micro molecular phenomena of the process which can not be measured. Model sequence: Intermediate Blocking, Complete Blocking, Cake Filtration Cross Flow.

Experiment 2

In experiment 2 the predictions calculated with a single model were not able to reproduce the data at all. Also the complete blocking model fails predicting the behaviour of the process. The presented approach fails also to show a continuous curve, nevertheless LSE = $3{,}81e^{-11}$ is more than ten times smaller. Although the optical run of the curve has not a very aesthetic appearance, a much better description of the process was presented by this method. Model sequence: Intermediate Blocking, Complete Blocking, Intermediate Blocking.

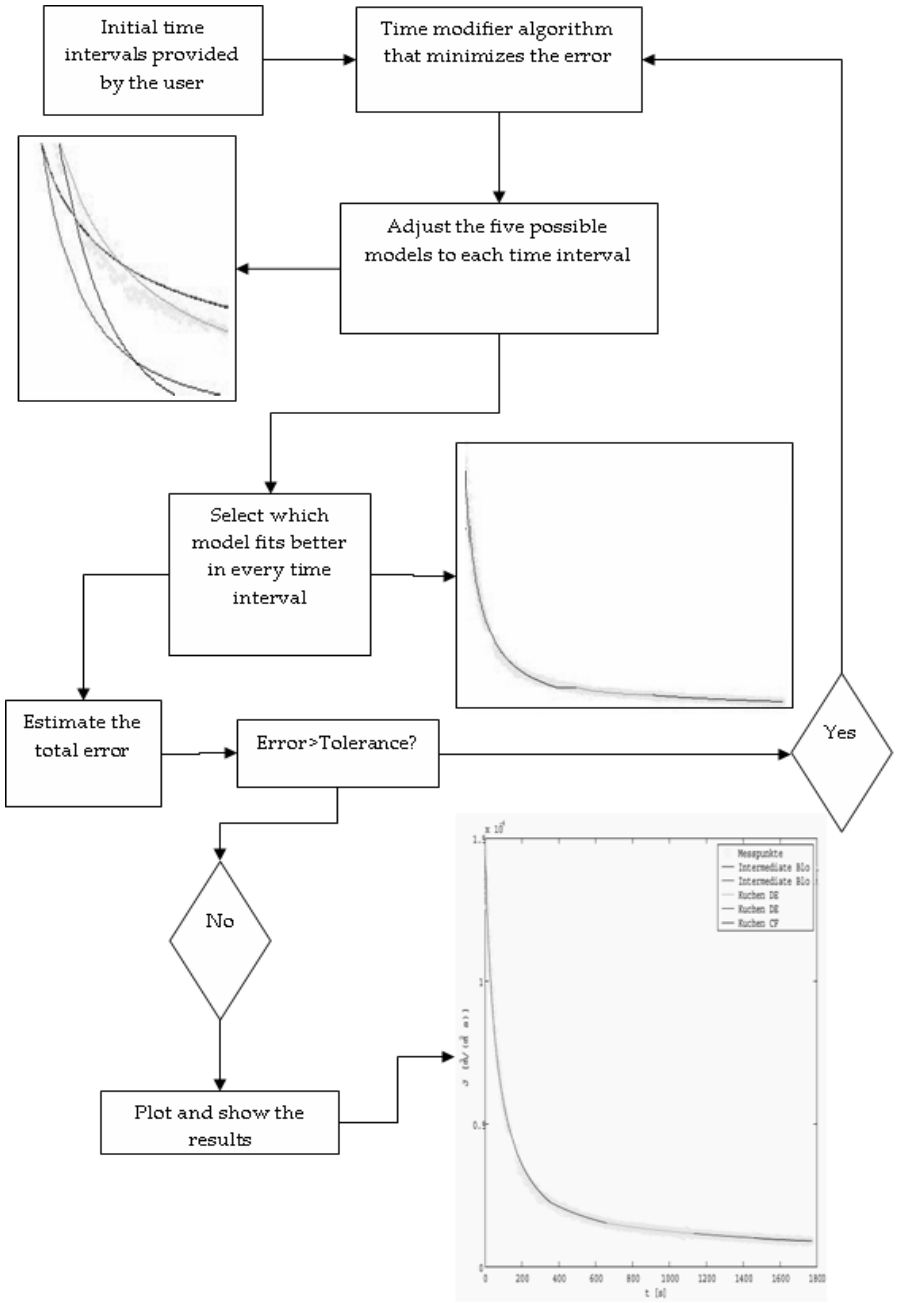

Fig. 5. Computational Strategy [16]

Rys. 5. Procedura obliczeniowa [16]

5. Conclusions

An Approach to Mechanistic Identification is presented which represents a useful tool to simulate complex and dynamical processes. It can be applied in any field where conventional models fail to predict the complete process by applying different models at different stages of the process. Additional to an improved simulation, the proposed framework provides new information about the process itself. Based on the analysis of the model sequence, different conclusions concerning the process itself and its operation can be concluded. Considering that the applied models are correct and the variance of the measurements is acceptable, the approach to mechanistic identification is able to determine the predominant phenomena occurring in the current interval.

In the case study of the MBR, the state of the membrane could be predicted such that counter current measures can be taken based just on data analysis. The information of the model applied is used to determine the state of the membrane. This information is useful to select the best of the three possible regenerative procedures.

Symbols

Φ	– objective function	
p_{es}	– estimated paramter	
t_k	– time at point k	[s]
$Z_{i,k}$	– measured value	
$\sigma_{i,k}$	– variance	
$C(t_k)$	– covariance matrix of error	
$C1$	– constant	
F	– fisher matrix	
p	– parameter vector	
C_M	– covariance matrix measure error	

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