

A Cognitive Approach to Develop Dynamic Models: Application to Polymerization Systems

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ABSTRACT: An alternative procedure based on cognitive approach is applied to develop dynamic models. The solution copolymerization of methyl methacrylate and vinyl acetate in a continuous stirred tank reactor is analyzed to illustrate the cognitive model development. Factorial planning was used to discriminate the process variables with higher impact on the process performance (effects) and they are used to built-up a dynamic model based on the functional fuzzy relationship of Takagi–Sugeno type. Gaussian membership functions are considered for the cognitive sets and subtractive clustering method supplied the parameters of the premises of the model. Consequence functions are obtained through an

optimization problem solved by a least square based algorithm. The kinetic parameters and reactor operating conditions are obtained from the literature and a mathematical model is considered as plant for identification data generation. Dynamic cognitive models showed satisfactory predictive capabilities and may be an interesting alternative to attack problems of modeling in chemical processes. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 981–992, 2007

Key words: fuzzy dynamic model; model identification; Takagi–Sugeno models; factorial planning; solution copolymerization

INTRODUCTION

The polymerization processes have a sufficiently complex behavior, which is characterized for nonlinear dynamic, and many times, it is impracticably a reliable theoretic model to be shaped. As a consequence, precise dynamic models of the process become increasingly difficult to be derived and solved as system complexity increases. This may lead to limitations and difficulties in the development and implementation of control strategies in chemical plants, especially when polymerization reactors are considered. However, nowadays, the requirements for high and safe operational performance together with the need to achieve the product

with desired quality demand impose that the plant to be operated under control.

Full detailed deterministic models are not tailored to be used as an internal control model. Thus, simplified models are formulated, with consequent restrictions in some phenomena representation. This may cause limitations on how the mathematical models tracks the process dynamic behavior with direct impact in the success of control strategies. Because of the great difficulties in modeling of polymerization processes, many works in literature report these difficulties. Bearing this in mind, it is clear that alternatives approaches are welcome as the proposed in this work.

Usually, some simplifications are made to become the problem easier to be formulated and solved. For instance, Embiruçu¹ and Embiruçu et al.² developed a dynamic mathematical model to describe the continuous process of polymerization of the ethylene with soluble Ziegler–Natta catalyzed in a set of stirred tank (CSTR) and tubular reactors. The tubular reactors had been assumed as PFR (plug flow reactor), and difficulties aroused for the resolution of set of partial differential equations of the PFR. The characteristics method was used, considering the similarity principle of the PFR with a batch reactor, which had simplified the numerical procedure. Only one type of active center was considered with objectives

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to simplify the model solution, despite the authors believe in the theory of multiplicity of active centers. Zeaiter et al.³ had developed a dynamic model for the polymerization in emulsion of the styrene in a half-batch reactor to foresee the product particle size distribution (PSD) and the molecular weight distribution (MWD). The developed model presented good results when compared with experimental data. Chen and Liu⁴ had developed a comprehensive mathematical model for single particle propylene polymerization mainly extended from polymeric multi-grain model (PMGM) and multigrain model (MGM) to describe kinetic behavior, MWD, monomer concentration, degree of polymerization and polydispersity index (PDI) for slurry-phase propylene polymerization using heterogeneous Ziegler–Natta catalysts. Further, special attention was also paid to discuss the computational effort, which is the most disadvantage of MGM. It has been shown that the significant computational time saving is also acquired by employing the novel solution methodology. Devadoss et al.⁵ had introduced and investigated a mathematical model for a free-radial frontal polymerization system. It was studied that a “pure” thiolene system that proceeds via a step-growth mechanism followed by a chain-transfer reaction. The model tracked the evolution of the species and the energy balance in the system. Both approximate analytical and full numerical solutions of the problem are given, and compared with the experimental results. Pontes⁶ developed a dynamic mathematical model to describe the continuous process of copolymerization of the ethene/1-butene with soluble Ziegler–Natta catalyzed in a set of tubular and agitated tank reactors. The author experienced difficulties in estimating the kinetic parameters, because of the large number of parameters to be identified. To overcome the problem, the parameters for the ethylene homopolymerization were estimated whereas for the copolymerization cross reactions, relationships from literature were used. Only one type of active center was considered. Clearly, such assumptions may lead to the lack of important process information.

Thus, the attainment of efficient more accurate representations for polymerization systems is necessary. Moreover, it is necessary to have a model, which should be at the same time easy and quick to solve as well as robust enough to capture the main process dynamics to develop a suitable control strategy.

In this work, a cognitive approach based on fuzzy concepts is proposed, considering both linguistic and mathematical functional representation. The approach allows to take into account both quantitative and qualitative informations, which lead the mathematical representation to accommodate the main process features. It is important to point out that the proposed approach has advantages when compared

with artificial neural network since, beyond the nonlinear behavior, qualitative information are also considered in the model building. Some works are presented in literature having related application of the fuzzy modeling in chemical processes, showing the potential of such approach. Alexandridis et al.⁷ introduced new systematic methodology to the problem of nonlinear system identification based on fuzzy systems. The proposed methodology was of general use and results in both a linguistic and an analytical model of the system. The method was successfully tested in the identification of certain operating regions in a Continuous Stirred Tank Reactor (CSTR) exhibiting various types of nonlinear behavior, such as limit cycles and multiple steady states. Accord to Abdelazim and Malik,⁸ fuzzy models are able to approximate any real continuous function up to a chosen accuracy, through the use of an algorithm for real-time identification of nonlinear systems using Takagi–Sugeno’s fuzzy models. A Takagi–Sugeno fuzzy system is trained incrementally each time step and is used to predict one-step ahead system output. The ability of the proposed identification to capture the nonlinear behavior of a synchronous machine is illustrated. Sala et al.⁹ detach the current research devoted to modeling and control through fuzzy methods, which presented good potential to be used in complex systems. In this way, fuzzy modeling methods appear as an alternative to solve modeling problems in polymerization reactions. In fact, they are quite attractive in terms of time and easiness to implementation.

Thus, this work presents the development of a fuzzy cognitive model to a copolymerization process. A deterministic mathematical model is used as plant for generation of the dynamic information and is assumed to be an adequate representation of the system. It will be shown that fuzzy model is able to reproduce the transient responses of deterministic model. The cognitive model was transformed in Fortran 90 software, which is portable and easy to use.

FUZZY LOGIC AS A MODELING TOOL FOR COMPLEX SYSTEMS

Most real world scheduling problems are very complex. Generating an optimal schedule is usually a time-consuming process, which requires various simplifying assumptions. Whether the schedule is generated by computationally demanding mathematical model or by a human expert, an appealing approach would be to devise a procedure to capture and model the underlying decision-making mechanism.¹⁰ Zadeh¹¹ considered a linguistic approach effective and versatile in modeling ill-defined systems with fuzziness or fully defined systems with realistic

approximations. Zadeh's work had a profound influence on the thinking about uncertainty because it challenged not only probability theory as the sole representation for uncertainty, but the very foundations upon which probability theory was based: classical binary (two-valued) logic.¹²

A fuzzy set contain elements that have varying degrees of membership in the set. This idea is in contrast with classical, or crisp, sets because members of a crisp set would not be members unless their membership was full, or complete, in that set. Elements in a fuzzy set, because their membership need not be complete, can also be members of other fuzzy sets on the same universe. All information contained in a fuzzy set is described by its membership function. These can be of various forms, such as Gaussian and triangular. The algorithms developed in this work incorporate Gaussian membership functions $\mu(x)$ for the inputs x , given by eq. (1):

$$\mu(x_i) = \exp\left[-\frac{1}{2}\left(\frac{x_i - c_i}{\sigma_i}\right)^2\right] \quad (1)$$

In eq. (1), x_i is the i th input variable, c_i is the i th center of the membership function (where the membership function achieves a maximum value), and σ_i is a constant related to spread of the i th membership function.

Fuzzy set properties and operations

Fuzzy sets follow the same properties as crisp sets, such as commutativity, associativity, distributivity, idempotency, identity, transitivity, and involution.¹³

The standard fuzzy operations are union, intersection, and complement, which are the same as those for classical sets when the range of membership values is restricted to the unit interval. This is an important information to built-up the knowledge basis for the process. However, these standard fuzzy operations are not the only operations that can be applied to fuzzy sets. For each of the three standard operations, there exists a broad class of functions whose members can be considered fuzzy generalizations of the standard operations. In such case, fuzzy intersections and fuzzy unions are usually referred to, in the literature, as *t-norms* and *t-conorms* (or *s-norms*), respectively. These *t-norms* and *t-conorms* are so named because they were originally introduced as triangular norms and triangular conorms, respectively, in study of statistical metric spaces.¹³ Table I indicates the more used *t-norms* and *t-conorms*, in relation the two fuzzy sets X_1 and X_2 , with elements x_1 and x_2 , respectively. The probabilistic *t-norm* and *t-conorm* will be applied in the calculation of the inferred exit of the fuzzy rule-base for the copolymerization process considered in this work.

TABLE I
Main *t-Norms* and *t-Conorms*

<i>t-Norm</i>	<i>t-Conorm</i>	Type
Min (x_1, x_2)	Max (x_1, x_2)	Zadeh
x_1x_2	$x_1 + x_2 - x_1x_2$	Probabilistic
Max ($x_1 + x_2 - 1, 0$)	Min ($x_1 + x_2, 1$)	Lukasiewicz
$\begin{cases} x_1, & \text{if } x_2 = 1 \\ x_2, & \text{if } x_1 = 1 \\ 0, & \text{if not} \end{cases}$	$\begin{cases} x_1, & \text{if } x_2 = 0 \\ x_2, & \text{if } x_1 = 0 \\ 1, & \text{if not} \end{cases}$	Weber

The inference procedure is described in the next topic.

Fuzzy modeling

The fuzzification, inference, and defuzzification stages must be processed for attainment of fuzzy models.

Fuzzification is the process of making a crisp quantity fuzzy. This can be done by simply recognizing that many of the quantities that are considered to be crisp and deterministic are actually not deterministic at all: they carry considerable uncertainty. If the form of uncertainty happens to arise because of imprecision, ambiguity, or vagueness, then the variable is probably fuzzy and can be represented by a membership function.¹⁴ These concepts are interesting to incorporate operator information on the process behavior and operation conditions in the model development. Possible changes in kinetic as well as heat and mass transfer parameters because of alterations in operating conditions may also be incorporated in the process model through the fuzzy approach.

The inference procedure is carried through by the expression of the type:

IF premise (antecedent),
THEN conclusion (consequent) (2)

This form is commonly referred to as the IF-THEN rule-based form; it is generally referred as the *deductive* form. It typically expresses an inference that may be used to express a fact (premise, antecedent, and hypothesis) in another fact named conclusion (consequent).

The defuzzification methods are used for the attainment of an effective action from the fuzzy model. In other words, defuzzification is the procedure of conversion of a fuzzy quantity to a precise quantity, just as fuzzification is the conversion of a precise quantity to a fuzzy quantity.

TAKAGI-SUGENO FUZZY MODEL

With the development of fuzzy systems, some fuzzy model systems design methods have appeared in fuzzy modeling field. Among various kinds of fuzzy modeling methods, Takagi and Sugeno¹⁵ proposed a design and analysis method for overall fuzzy systems, in which the qualitative knowledge of a system was first represented by a set of local Takagi–Sugeno fuzzy model. In this approach, the Takagi–Sugeno fuzzy model (TS) substitutes the consequent fuzzy sets in a fuzzy rule by a linear equation of the input variables. Local dynamics in different state-space regions are represented by linear models and the overall model of the system is represented as the interpolation of these linear models. Therefore, it has a convenient dynamic structure so that some well-established linear systems theory can be easily applied for theoretical analysis and design of the overall closed-loop system.¹⁴

The TS method was proposed in an effort to develop a systematic approach to generate fuzzy rules from a given input–output data set. A typical rule in a TS model, which has two-inputs x_1 and x_2 , and output y , has the form:

$$\text{IF } x_1 \text{ is } X_1 \text{ and } x_2 \text{ is } X_2, \text{ THEN } y \text{ is } y = f(x_1, x_2) \quad (3)$$

where X_1 and X_2 are fuzzy sets (membership functions) of x_1 and x_2 , respectively, and $y = f(x_1, x_2)$ is a crisp consequent function. The generalization of expression (3) for a number of entrances n leads to the TS model as follow:

If (x_1 is $X_{i,1}$) **and** (x_2 is $X_{i,2}$) **and** ... **and** (x_j is $X_{i,j}$)

and ... **and** (x_n is $X_{i,n}$) **then**

$$y_i = a_{i1}x_1 + a_{i2}x_2 + \cdots + a_{ij}x_j + \cdots + a_{in}x_n \quad (4)$$

where $i = 1, \dots, R$, being R the rules number of the fuzzy model; $j = 1, \dots, n$; and a_{ij} are parameters of the consequent function of the fuzzy model.

For the system analyzed in this work, the subtractive clustering method is used for determination of the rules number and parameters of the membership functions of the antecedent part of the fuzzy model. The algorithm forms rules (or clusters) with training data using a nearest neighbor approach for the fuzzy system. Consequence functions parameters are obtained through an optimization problem solved by a least square based algorithm, through error minimization between the plant data and the foreseen values for the fuzzy model. Details on subtractive clustering and least square methods are given by Chiu^{16,17} and Passino and Yurkovich,¹⁸ respectively.

The inferred numerical exit is defined by the weighed average of referring numerical exits i to each rule, calculated by:

$$y = \frac{\sum_{i=1}^R f_i \mu_i(x)}{\sum_{i=1}^R \mu_i(x)} \quad (5)$$

where $\mu_i(x)$ are membership functions and f_i is a consequent function to each rule i .

This is the basic procedure proposed in this work to develop a fuzzy model.

IDENTIFICATION OF FUZZY DYNAMIC MODELS

It is important to draw attention that a series of decisions must be taken in the initial phases of the modeling process, which will influence directly in the quality of the obtained model.

A first point to be considered is the definition of the fuzzy structure model that composes the system base of rules. The variables that will be used as well as the form of interconnection among them (if the relation is linear or non linear, being able to be exponential, power, mixing, among others) must be defined. The number and types of the chosen variables must be in accord to the problem necessity. These models will possess a dynamic configuration in such a way to represent the behavior of the process throughout a time horizon.

In fact, the fuzzy models generally use the last information in their structures and use last values of the chosen exits as the entrance of the model. The number of last variable information used to construct the model is an important parameter in the performance of the model. In fact, it can be seen as an optimization topic that must be taken into account in the model construction.

Defined the structure, the next stage is the data generation for the identification of the model. In this point, the maximum and minimum limits of variation of the variables must be chosen, so that the model operation range is determined, considering the desired objectives. Initially, the training data are generated, which are used for attainment the parameters of the model. This model is validated later through the application of the test data. The data generation is carried out through the excitement of the entrance variables of the system, being able to be basically of two forms: random, or through an experiment planning.

The procedure of the model parameters determination involves matrix inversion. Thus, it is important to detach that the choice of the values of frequency is of extreme importance; therefore, a reduced excitement can cause problems of matrix

ill-conditioning, with consequent problem in the inversion procedure. On the other hand, a maximum excitement frequency able to shape the stationary values must be guaranteed. In relation to the amplitude of excitement, it has to be emphasized that this data must take in account the operational range of the model. The training and test data generation is carried out in different excitement conditions of the entrance variables in terms of frequency and amplitude.

Another important point to be considered in the dynamic fuzzy models development is the determination of the sampling rate. In such case, it has to be taken into account the time constant of the process preventing at the same time problems of matrix conditioning when it is too large. On the other hand, for the case of the model for control of processes, the value of the sampling rate must be related with the controller action interval, so that very small values lead to improper control actions.

MODEL VALIDATION

In this work, the results of the model validation are illustrated through figures and quantified through of the "average quadratic error," given by eq. (6):

$$\text{error} = \sqrt{\frac{\sum_{k=1}^m (\bar{y}_k - y_k)^2}{m}} \quad (6)$$

where k is the considered time instant, m is the number of considered discrete instants, \bar{y}_k is the predicted exit by the fuzzy model in instant k , and y_k is the exit of the process in instant k (deterministic model).

CASE STUDY

The process considered in this work as case study is the free radical solution copolymerization of methyl methacrylate and vinyl acetate in a continuous stirred tank reactor.¹⁹

Figure 1 is a flow diagram of a copolymerization reactor with a recycle loop. Monomer A is methyl methacrylate, monomer B is vinyl acetate, the solvent is benzene, the initiator is azobisisobutyronitrile (AIBN) and the chain transfer agent is acetaldehyde. The monomer stream may also contain inhibitors such as *m*-dinitrobenzene (*m*-DNB).

Monomers A and B are continuously added with initiator, solvent, and chain transfer agent. In addition, an inhibitor may enter with the fresh feeds as an impurity. These feed streams are combined (Stream 1) with the recycle stream (Stream 2) and flow to the reactor (Stream 3), which is assumed to be a jacketed, well-mixed tank. A coolant flows through the jacket to remove the polymerization heat. Polymer, solvent, unreacted monomers, initiator, and chain transfer agent flow out of the reactor to the separator (Stream 4). Here, polymer is re-

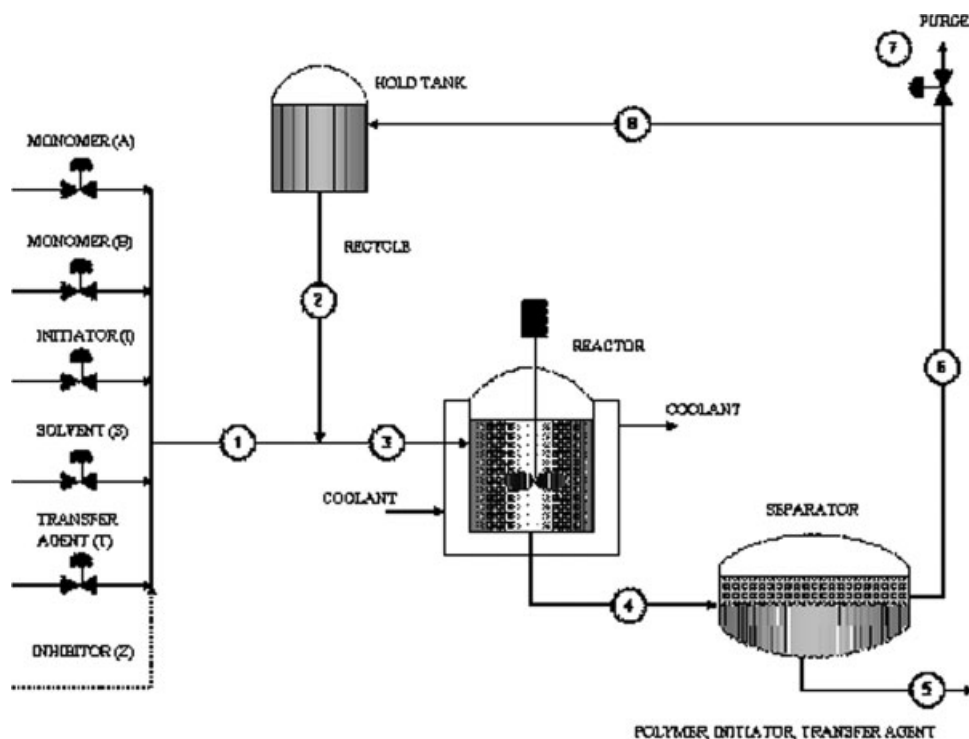


Figure 1 Basic process configuration.

TABLE II
Steady-State Operating Conditions

Inputs	
Monomer A (MMA) feed rate	$G_{af} = 17$ kg/h
Monomer B (VAc) feed rate	$G_{bf} = 90$ kg/h
Initiator (AIBN) feed rate	$G_{if} = 0.18$ kg/h
Solvent (benzene) feed rate	$G_{sf} = 36$ kg/h
Chain transfer (Acetaldehyde) feed rate	$G_{tf} = 2.7$ kg/h
Inhibitor (<i>m</i> -DNB) feed rate	$G_{zf} = 0$
Reactor jacket temperature	$T_j = 336.15$ K
Reactor feed temperature	$T_{rf} = 353.15$ K
Purge ratio	$\xi = 0.05$
Reactor parameters	
Reactor volume	$V_r = 1$ m ³
Reactor heat transfer area	$S_r = 4.6$ m ²
Outputs	
Polymer production rate	$G_{pi} = 23.3$ kg/h
Mole fraction of A in polymer	$Y_{ap} = 0.56$
Weight-average molecular weight	$M_{pw} = 35000$ kg/kmol
Reactor temperature	$T_r = 353.01$ K

moved from the stream (Stream 5). Residual initiator and chain transfer agent are also removed in this step. In real process, the separator often involves a series of steps, which may include dryers and distillation columns. Here, unreacted monomers and solvent (Stream 6) continue on to a purge point (Stream 7), which represents venting and other losses. Purging is required to prevent the accumulation of inerts in the system. After the purge, the monomers and solvent (Stream 8) are stored in the recycle hold tank, which acts as a surge capacity to smooth out variations in the recycle flow and composition. The effluent (Stream 2) recycle is then added to the fresh feeds.

The important reactor output variables for product quality control are the polymer production rate (G_{pi}), mole fraction of monomer A in the copolymer (Y_{ap}), weight-average molecular weight (M_{pw}), and reactor temperature (T_r). The inputs are the reactor flows of monomer A (G_{af}), monomer B (G_{bf}), initiator (G_{if}), chain transfer agent (G_{tf}), solvent (G_{sf}), inhibitor (G_{zf}), the temperature of the reactor jacket (T_j), and the temperature of the reactor feed (T_{rf}). The reactor, separator and hold tank contents at startup are pure solvent preheated to 353.15 K.

The steady-state operating point is summarized in Table II. Under these conditions, the reactor residence time is $\theta_r = 6$ h and the overall reactor monomer conversion is 20%. These operating conditions ensure that the viscosity of the reaction medium remains moderate. Table II also indicates that the temperature of the reactor feed T_{rf} is practically equal to the reactor temperature T_r , because this work was chosen to simulate reactor operation with a preheated feed where the source of heat removal is through the jacket.

Feedforward control of recycle

The presence of the recycle stream introduces disturbances in the reactor feed, which affects the polymer properties. In this way, Congalidis et al.¹⁹ implemented a feedforward controller in the process to compensate for these disturbances by manipulating the fresh feeds to maintain constant feed composition and flow to the reactor. Feedforward control of the recycle stream enabled the designer to separate the control of the reactor from the rest of the process. Thus, the reactor can be separately analyzed.

The feedforward control equations were obtained by writing component balances around the recycle addition point. For example, the mole balance for monomer A is:

$$F_{a3} = F_{a1} + y_{a2}F_2 \quad (7)$$

Equation (7) is then solved for the fresh feed of monomer A since it is desired to keep the goal flow of monomer A to the reactor (F_{a3}) constant:

$$F_{a1} = F_{a3} - y_{a2}F_2 \quad (8)$$

Since only monomers A and B and solvent are present in the recycle, only these three components have feedforward control equations. The corresponding equations for fresh feeds of monomer B and solvent are:

$$F_{b1} = F_{b3} - y_{b2}F_2 \quad (9)$$

$$F_{s1} = F_{s3} - y_{s2}F_2 \quad (10)$$

If any feedforward control equation causes a fresh feed to go negative, the value of that fresh feed is set to zero.

Deterministic model

The analyzed copolymerization process has described in literature a deterministic mathematical model and kinetic parameters, which is considered as plant for data generation for the identification of the cognitive model. This is composed by a set of algebraic and ordinary differential equations and was chosen for already being a developed and validated model. Besides, such system presents complex dynamic behavior with highly nonlinear characteristics. Details on the deterministic model are given in Congalidis et al.¹⁹ and Maner and Doyle.²⁰

Discriminate of the process variables

The analyzed system consists of seven entrances (G_{af} , G_{bf} , G_{if} , G_{tf} , G_{sf} , and T_j) and four exits (G_{pi} , Y_{ap} , M_{pw} , and T_r). The temperature of the reactor feed T_{rf} was considered constant and purge ratio is under action of

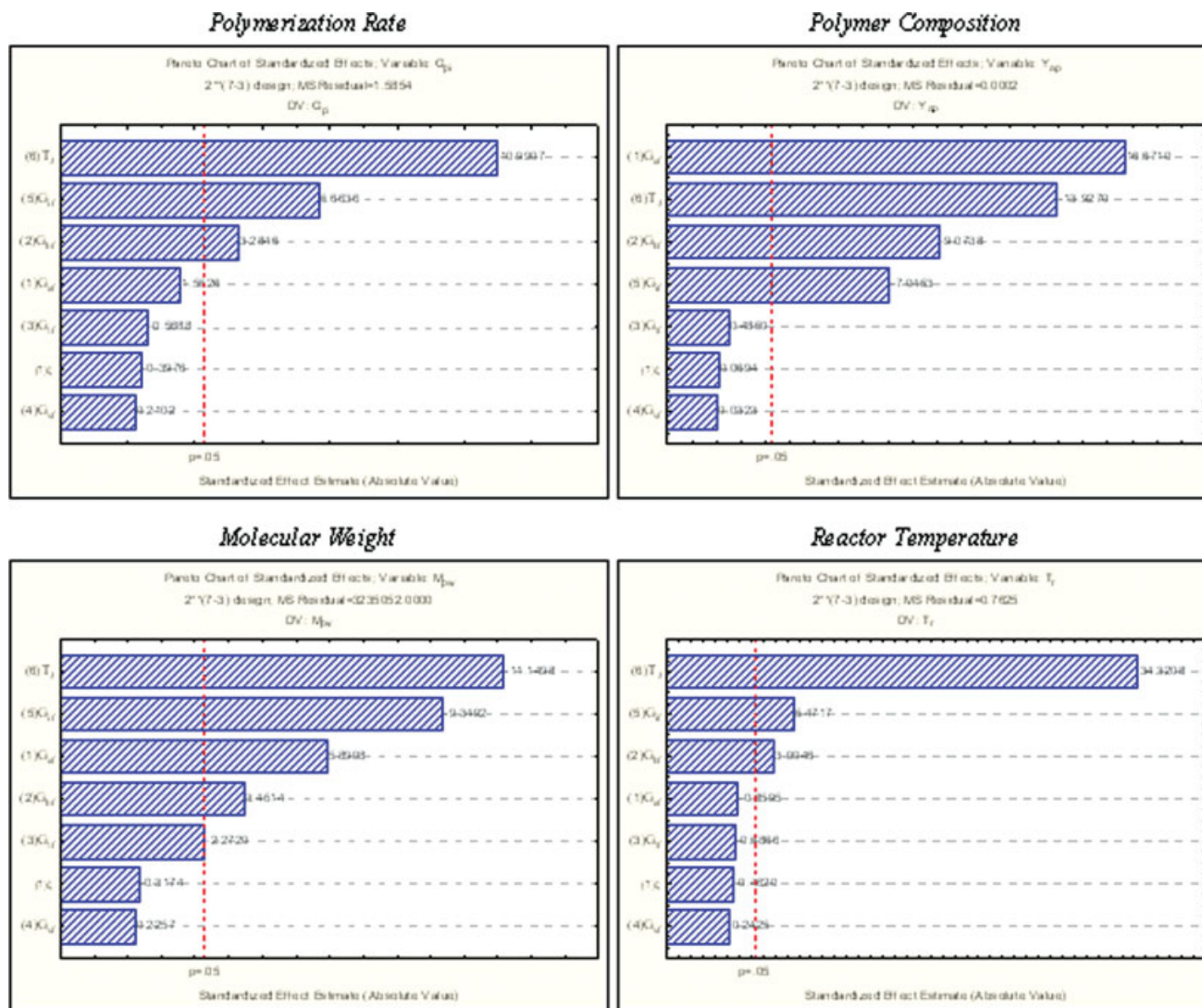


Figure 2 Pareto chart to output variables. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the feedforward controller. Table II also indicates that inhibitor feed rate is equal the zero. Thus, factorial planning using *Statistica Version 7.0 Software* was used to discriminate the process variables with higher impact on the process performance (effects). They are used to build up a dynamic model based on the functional fuzzy relationship of Takagi–Sugeno type. The deterministic mathematical model of the process proceeding from literature was solved by a Runge–Kutta algorithm type in a software written in FORTRAN 90 (Compaq Visual Fortran 6.6) language. The initial concentrations and operational conditions in the entrance of the reactor had been gotten from Maner and Doyle.²⁰ This software was used in the data generation of exit for each set of entrance defined by experimental planning through the *Statistica Version 7.0 Software*. Figure 2 illustrates the Pareto chart of standardized effects to four exits. Table III summarizes the results.

Functional dynamic models

As already described, the functional fuzzy models developed in this work are of the type Takagi–Sugeno, whose structures were previously defined [eq. (4)]. The next stage for the models construction is the attainment of the data for process identification. It is clear that the data set of training must represent the process in the best way it is possible, therefore this is a decisive factor to achieve a good

TABLE III
Discrimination of the Process Variables

Outputs	Variables of higher impact
G_{pi}	T_j, G_{if}, G_{bf}
Y_{ap}	$G_{af}, T_j, G_{bf}, G_{if}$
M_{pw}	$T_j, G_{if}, G_{af}, G_{bf}$
T_r	T_j, G_{if}, G_{bf}

TABLE IV
Discrimination of the Variation Intervals

Input variables	Variation intervals (%)
G_{af}	± 15
G_{bf}	± 15
G_{if}	± 15
T_j	± 3

model. The output variable data were obtained through random changes in the input variables with higher impact on the process performance. The deterministic model was used in this task. The considered ranges for each input variable are presented in Table IV, being related to the steady-state values. In this way, identification data of the process (the input/output set) necessary for the generation of

cognitive model were obtained. Figure 3 presents the identification data for each one of the output variables. It was used as a sampling rate of 1 h and a simulation interval of 400 h. From the identification data, algorithms for generation of the cognitive models were build up. They are used to develop the dynamic model based on the functional fuzzy relationship of Takagi–Sugeno type. Gaussian membership functions were used for the fuzzy sets and subtractive clustering method supplied the parameters of the premises of the model and rules number. Consequence functions were obtained through an optimization problem solved by a least square based algorithm. Tables V–VIII show the parameters for the fuzzy models. The current value and the last value to each input variable as well as the last value

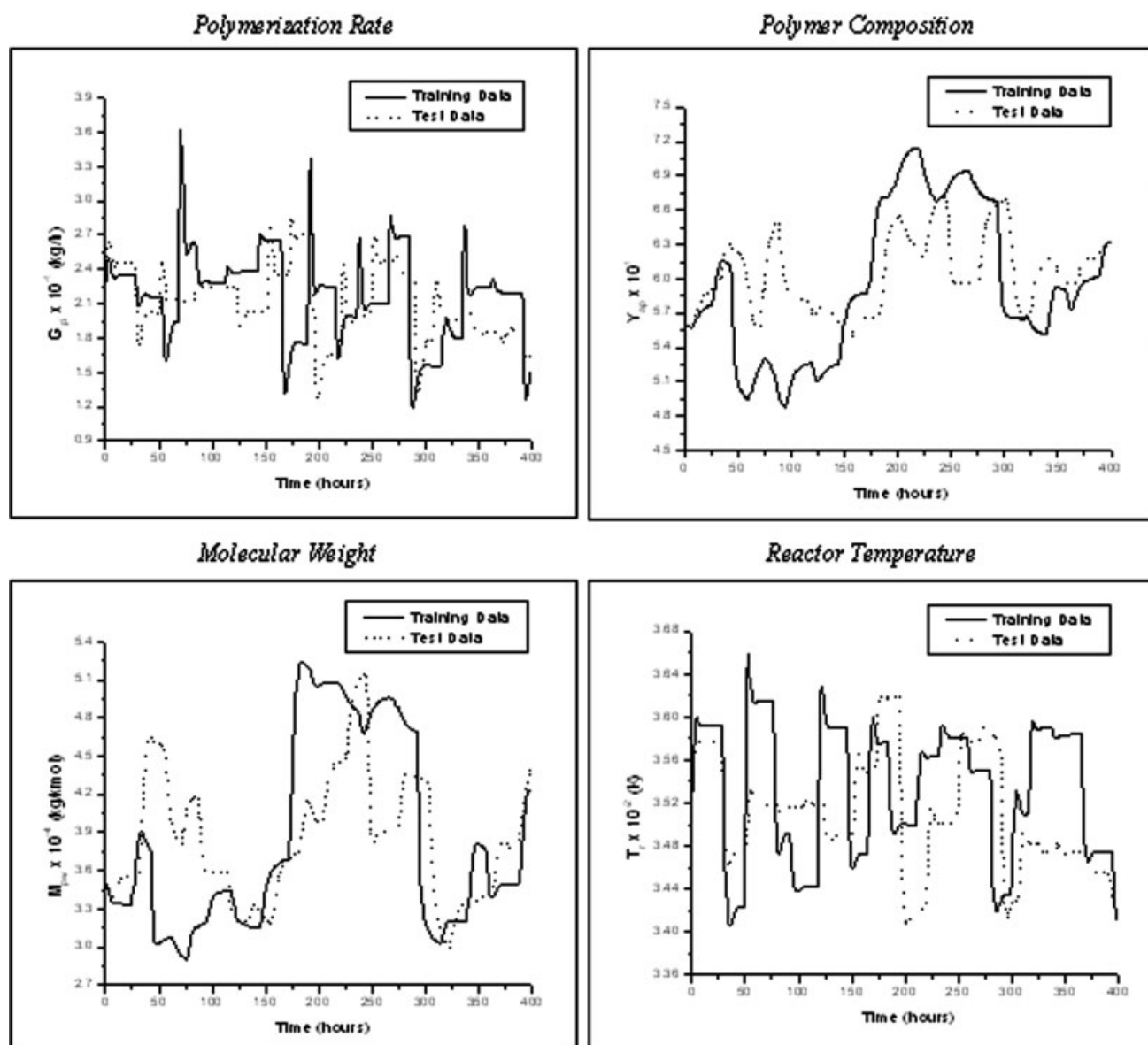


Figure 3 Identification data to output variables.

TABLE V
Parameters to Polymer Production Rate (G_{pi})

Rules number = 2							
Antecedent part							
Rule 1		Rule 2		Rule 1		Rule 2	
$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$
$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$	
86.59	15.78	8.91	15.78	86.59	15.78	8.91	15.78
$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$	
7.42	16.69	1.11	16.69	7.42	16.69	1.11	16.69
$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$	
38.44	17.09	84.90	17.09	38.44	17.09	84.90	17.09
$y_1(k - 1)$		$y_1(k - 1)$		$y_1(k - 1)$		$y_1(k - 1)$	
50.15	19.70	13.80	19.70	50.15	19.70	13.80	19.70
Consequent part							
Rule i	$a_{i1} \times 10^2$	$a_{i2} \times 10^2$	$a_{i3} \times 10^2$	$a_{i4} \times 10^2$	$a_{i5} \times 10^2$	$a_{i6} \times 10^2$	$b_{i1} \times 10^2$
$i = 1$	26.64	-17.36	4.06	1.26	3.69	-3.29	84.01
$i = 2$	48.12	-20.88	4.12	4.25	9.96	-5.40	53.45

of output variable were considered as entrances for the cognitive model.

In the Tables V–VIII, k refers to time instant, and $u_1, u_2, u_3,$ and u_4 refer to $T_j, G_{af}, G_{bf},$ and $G_{if},$ respectively; y_j is the j th output variable; a_{in} and b_{ij} are consequence functions parameters of the input variables and j th output variable, respectively, where $i = 1, \dots, R$ (rules number) and n is the number of parameters of the entrances.

The i th rule to each exit is shown in eqs. (11–14). A_{in} and B_{ij} are the membership functions of entrances and exit, respectively.

TABLE VI
Parameters to Fraction of Monomer A in the Copolymer (Y_{ap})

Rules number = 3									
Antecedent part									
Rule 1		Rule 2		Rule 3		Rule 1		Rule 2	
$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$
$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$	
23.49	16.22	86.59	16.22	8.02	16.22	23.49	16.22	86.59	16.22
$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$	
60.04	16.66	18.89	16.66	51.79	16.66	60.04	16.66	18.89	16.66
$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$	
18.57	17.47	38.44	17.47	95.01	17.47	18.57	17.47	38.44	17.47
$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$	
4.52	16.07	7.42	16.07	76.01	16.07	4.52	16.07	7.42	16.07
$y_2(k - 1)$		$y_2(k - 1)$		$y_2(k - 1)$		$y_2(k - 1)$		$y_2(k - 1)$	
45.60	17.43	86.81	17.43	12.06	17.43	45.60	17.43	86.81	17.43
Consequent part									
Rule i	$a_{i1} \times 10^2$	$a_{i2} \times 10^2$	$a_{i3} \times 10^2$	$a_{i4} \times 10^2$	$a_{i5} \times 10^2$	$a_{i6} \times 10^2$	$a_{i7} \times 10^2$	$a_{i8} \times 10^2$	$b_{i2} \times 10^2$
$i = 1$	0.17	2.43	0.47	-2.32	3.60	-0.21	0.75	0.31	97.30
$i = 2$	1.89	2.30	-2.72	-2.09	0.80	-0.63	-0.75	-0.69	97.72
$i = 3$	-6.95	3.33	0.96	-3.74	0.54	0.29	4.20	-1.92	97.75

TABLE VII
Parameters to Weight-Average Molecular Weight (M_{pw})

Rules number = 3									
Antecedent part									
Rule 1		Rule 2		Rule 3		Rule 1		Rule 2	
$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$
$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$	
60.04	16.66	8.91	16.66	60.04	16.66	8.91	16.66	95.55	16.66
$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$	
4.52	16.07	22.64	16.07	4.52	16.07	22.64	16.07	95.41	16.07
$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$		$u_2(k), u_2(k - 1)$	
23.49	16.22	84.90	16.22	23.49	16.22	84.90	16.22	63.39	16.22
$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$	
18.57	17.47	1.11	17.47	18.57	17.47	1.11	17.47	89.76	17.47
$y_3(k - 1)$		$y_3(k - 1)$		$y_3(k - 1)$		$y_3(k - 1)$		$y_3(k - 1)$	
25.00	17.60	93.85	17.60	25.00	17.60	93.85	17.60	7.04	17.60
Consequent part									
Rule i	$a_{i1} \times 10^2$	$a_{i2} \times 10^2$	$a_{i3} \times 10^2$	$a_{i4} \times 10^2$	$a_{i5} \times 10^2$	$a_{i6} \times 10^2$	$a_{i7} \times 10^2$	$a_{i8} \times 10^2$	$b_{i3} \times 10^2$
$i = 1$	2.48	-5.81	2.56	-0.79	-1.28	3.35	5.69	-0.52	93.50
$i = 2$	-12.18	13.71	-1.22	1.97	1.77	1.16	0.66	0.20	96.69
$i = 3$	-10.35	-11.23	15.97	0.88	2.19	4.97	-1.05	1.55	77.07

Output variable : G_{pi}

Rule i : IF ($u_1(k)$ is A_{i1}) and ($u_1(k - 1)$ is A_{i2}) and ($u_4(k)$ is A_{i3}) and ($u_4(k - 1)$ is A_{i4}) and ($u_3(k)$ is A_{i5}) and ($u_3(k - 1)$ is A_{i6}) and ($y_1(k - 1)$ is B_{i1}) THEN $y_{i1}(k + 1) = a_{i1}u_1(k) + a_{i2}u_1(k - 1) + a_{i3}u_4(k) + a_{i4}u_4(k - 1) + a_{i5}u_3(k) + a_{i6}u_3(k - 1) + b_{i1}y_1(k - 1)$ (11)

TABLE VIII
Parameters to Reactor Temperature (T_r)

Rules number = 3									
Antecedent part									
Rule 1		Rule 2		Rule 3		Rule 1		Rule 2	
$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$	$c_i \times 10^2$	$\sigma_i \times 10^2$
$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$		$u_1(k), u_1(k - 1)$	
83.45	15.82	10.74	15.82	83.45	15.82	10.74	15.82	81.43	15.82
$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$		$u_4(k), u_4(k - 1)$	
22.32	16.05	69.15	16.05	22.32	16.05	69.15	16.05	12.32	16.05
$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$		$u_3(k), u_3(k - 1)$	
43.58	16.23	23.27	16.23	43.58	16.23	23.27	16.23	89.85	16.23
$y_4(k - 1)$		$y_4(k - 1)$		$y_4(k - 1)$		$y_4(k - 1)$		$y_4(k - 1)$	
86.64	19.39	15.14	19.39	86.64	19.39	15.14	19.39	80.26	19.39
Consequent part									
Rule i	$a_{i1} \times 10^2$	$a_{i2} \times 10^2$	$a_{i3} \times 10^2$	$a_{i4} \times 10^2$	$a_{i5} \times 10^2$	$a_{i6} \times 10^2$	$a_{i7} \times 10^2$	$a_{i8} \times 10^2$	$b_{i4} \times 10^2$
$i = 1$	32.97	24.18	-2.50	5.95	1.52	5.42	39.36		
$i = 2$	21.52	35.15	1.85	-0.62	12.58	-10.26	47.44		
$i = 3$	40.87	10.06	-28.99	13.16	-32.37	31.52	52.48		

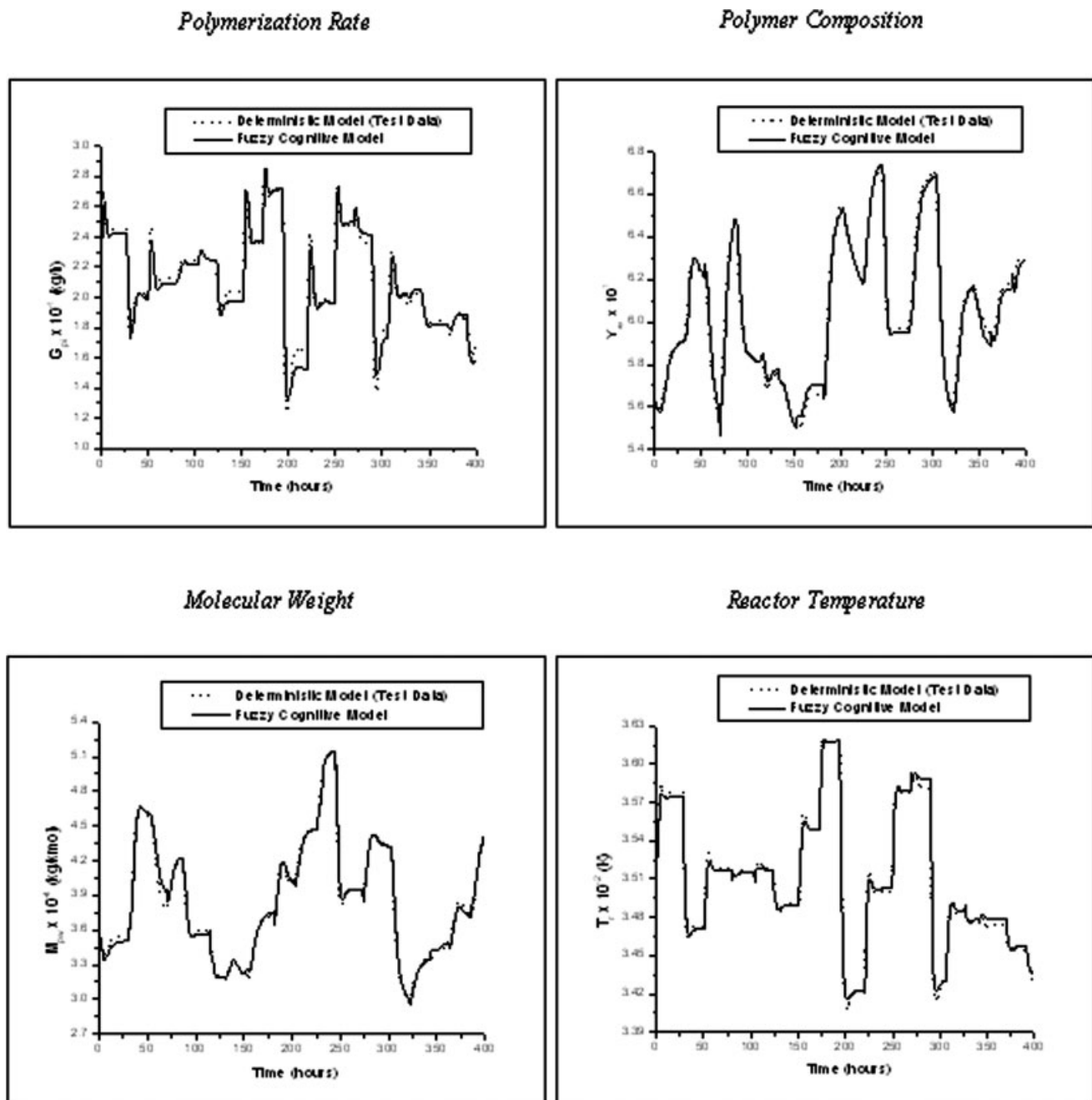


Figure 4 Validation of the fuzzy model to output variables.

Output variable : Y_{ap}

Rule i : **IF** ($u_2(k)$ is A_{i1}) and ($u_2(k-1)$ is A_{i2}) and ($u_1(k)$ is A_{i3}) and ($u_1(k-1)$ is A_{i4}) and ($u_3(k)$ is A_{i5}) and ($u_3(k-1)$ is A_{i6}) and ($u_4(k)$ is A_{i7}) and ($u_4(k-1)$ is A_{i8}) and ($y_2(k-1)$ is B_{i2}) **THEN** $y_{2i}(k+1) = a_{i1}u_2(k) + a_{i2}u_2(k-1) + a_{i3}u_1(k) + a_{i4}u_1(k-1) + a_{i5}u_3(k) + a_{i6}u_3(k-1) + a_{i7}u_4(k) + a_{i8}u_4(k-1) + b_{i2}y_2(k-1)$ (12)

Output variable : M_{pw}

Rule i : **IF** ($u_1(k)$ is A_{i1}) and ($u_1(k-1)$ is A_{i2}) and ($u_4(k)$ is A_{i3}) and ($u_4(k-1)$ is A_{i4}) and ($u_2(k)$ is A_{i5}) and ($u_2(k-1)$ is A_{i6}) and ($u_3(k)$ is A_{i7}) and ($u_3(k-1)$ is A_{i8}) and ($y_3(k-1)$ is B_{i3}) **THEN** $y_{3i}(k+1) = a_{i1}u_1(k) + a_{i2}u_1(k-1) + a_{i3}u_4(k) + a_{i4}u_4(k-1) + a_{i5}u_2(k) + a_{i6}u_2(k-1) + a_{i7}u_3(k) + a_{i8}u_3(k-1) + b_{i3}y_3(k-1)$ (13)

TABLE IX
Errors Quantification for Output Variables

Outputs	Average quadratic error [eq. (6)]
G_{pi}	0.6915
Y_{ap}	0.0040
M_{pw}	536.72
T_r	0.6645

Output variable : T_r

Rule i : **IF** ($u_1(k)$ is A_{i1}) and ($u_1(k-1)$ is A_{i2}) and ($u_4(k)$ is A_{i3}) and ($u_4(k-1)$ is A_{i4}) and ($u_3(k)$ is A_{i5}) and ($u_3(k-1)$ is A_{i6}) and ($y_4(k-1)$ is B_{i4}) **THEN** $y_{4i}(k+1) = a_{i1}u_1(k) + a_{i2}u_1(k-1) + a_{i3}u_4(k) + a_{i4}u_4(k-1) + a_{i5}u_3(k) + a_{i6}u_3(k-1) + b_{i4}y_4(k-1)$ (14)

Figure 4 presents the validation results of the fuzzy models for the test data set. As can be seen, a very good prediction was obtained for the four output variables. Table IX shows the result of the errors quantification for each exit. As observed, a very small average quadratic error for the four output variables was obtained, having as reference its respective dimensions.

CONCLUSIONS

Fuzzy cognitive models were developed in this work for a copolymerization process. The analysis of these models, together with results shown on Figure 4 and Table IX, allow to conclude that these models represent the process in a very satisfactory way for the four output variables. In fact, the use of fuzzy dynamic models represents a good alternative for modeling polymerization processes. The main advantage in such model approach is the use of input/output data set together with qualitative information. Also, the ability of fuzzy models to deal with uncertainties in kinetic and transfer parameters as well as the lack of complete information on the phenomena taking place in the system has to be considered. As pointed out, polymerization processes are usually complex and difficult to model and solve through deterministic mathematical modeling approach, especially to be used in control. A fuzzy model is simpler to be built-up, identified, and solved, and it appears to be a suitable way to model a system for control and real-time optimization.

NOMENCLATURE

A Monomer A, membership function of the input variable

AIBN Azobisisobutyronitrile
 B Monomer B, membership function of the output variable
 a Parameter of the consequent function of the fuzzy model refer to input variable
 b Parameter of the consequent function of the fuzzy model refer to output variable
 c Center of the Gaussian membership function
 CSTR Continuous stirred tank reactor
 F Molar flow rate (kmol/h)
 f Consequent function of the fuzzy model
 G Mass flow rate (kg/h)
 k Time instant
 M Molecular weight (kg/kmol)
 m Number of time instants
 MGM Multigrain model
 MWD Molecular weight distribution
 PDI Polydispersity index
 PFR Plug flow reactor
 PMGM Polymeric multigrain model
 PSD Particle size distribution
 R Rules number of the fuzzy model
 S Surface area (m²)
 T Temperature (K)
 u Input variable
 v Volume (m³)
 X Fuzzy sets (Gaussian membership function)
 x Input variable, elements of the fuzzy sets
 Y Mole fraction
 y Consequent function of the fuzzy model, process exit, output variable
 \hat{y} Predicted exit by the fuzzy model

Greek letters

μ Gaussian membership function
 θ Residence time (h)
 σ Constant related to spread of the Gaussian membership function
 ξ Molar purge fraction

Subscripts

a Monomer A
 b Monomer B
 f Feed to the reactor
 i Initiator, instantaneous, rule of the fuzzy model
 j Cooling jacket, entrance of the fuzzy model, output variable
 k Time instant
 n Number of entrances of the fuzzy model, input variable
 p Dead polymer
 r Reactor
 s Solvent
 t Chain transfer agent
 w Weight average polymer property
 z Inhibitor

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