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Temperature control adjustment in a batch polymerization reactor depending on the operating mode and the cooling fluid flow rate - Internal model control and exothermic reactions

Ajuste do controle de temperatura em um reator de polimerização em batelada dependendo do modo de operação e da vazão do fluido refrigerante – IMC e reações exotérmicas

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Abstract: The design of a temperature controller for batch polymerization processes present some challenges due to high heat generation and the increase of viscosity of the reaction medium typical of these reactions. This work exhibits the design and implementation of two types of temperature controllers for a batch reactor that uses water as a cooling fluid. A cooling tower and a boiler are used for cooling and/or heating the cooling fluid, respectively. To check the robustness of controlling the cooling fluid's flow rate was changed from one reaction to another. The transition between operational modes (isothermal and isoperibolic) and the insertion of reactants during the reaction was also subjected to analysis and deemed disturbances within the system. A model for the process from real data was estimated, and the transfer function of the controllers was set. In the case of PID structure (Proportional, Integral, and Derivative) the derivative action was disregarded because it requires a strong performance of automatic valves reducing its useful life span. The IMC (Internal Model Control) structure type ensured the system stability even in the presence of disturbances.

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Resumo: O projeto de um controlador de temperatura para processos de polimerização em batelada apresenta alguns desafios devido à alta geração de calor e ao aumento da viscosidade do meio reacional típico dessas reações. Este trabalho apresenta o projeto e a implementação de dois tipos de controladores de temperatura para um reator batelada que utiliza água como fluido refrigerante. Uma torre de resfriamento e uma caldeira são usadas para resfriar e/ou aquecer o fluido de resfriamento, respectivamente. Para verificar a robustez do controle, a vazão do fluido refrigerante foi alterada de uma reação para a outra. A mudança entre os modos de operação (isotérmico e isoperibólico) e alimentação de reagentes durante a reação também foram analisados e considerados como perturbações no sistema. Um modelo para o processo a partir de dados reais foi estimado e a função de transferência dos controladores foi definida. No caso da estrutura PID (Proporcional, Integral e Derivativo) a ação derivativa foi desconsiderada por exigir um forte desempenho das válvulas automáticas reduzindo sua vida útil. A estrutura do tipo IMC (Internal Model Control) garantiu a estabilidade do sistema mesmo durante a existência de perturbações.

Palavras-chave: Controle. IMC. PID. Reator. Temperatura.

1. INTRODUCTION

Polymer production with previously specified quality parameters requires continuous monitoring and process control to ensure product authenticity. As a result, calorimetry is one of many techniques used to monitor polymerization reactions Online. It measures energy exchanges during a system reaction (including phase change) under conditions that enables all process rates to be measured and analyzed.

Since calorimetry aims to determine the heat generated by a reaction, its use is extensive among chemical processes, particularly in essentially exothermic polymerization reactions. It can be said that its priority is temperature control to ensure safe operation. Industrial reactors have a small heat exchange area-to-volume ratio, leading to a relatively limited capacity to remove heat. Furthermore, together with the high enthalpy of polymerization reactions and the viscosities of latexes produced in many industrial processes, this relation can lead to situations that result in thermal runaway. An example is the accumulation of monomers in the reactor, which can lead to dangerous situations. This accumulation can occur due to different factors: varying monomer inhibition due to batch-to-batch variations, incorrect initiator level, poor temperature control, poor mixing in the reactor, or the presence of impurities. In such cases, a subsequent sudden heat release occurs when polymerization recommences. This process generates a poor-quality product (with wrong composition and low molecular weight) and, in extreme cases, thermal runaway. Online reaction calorimetry can prevent this potentially hazardous condition by rapidly detecting the decrease in the reaction heat resulting from monomer accumulation. This detection allows corrective measures such as reducing or stopping reagent addition (Kummer; Nagy, & Varga, 2020).

Laboratory reactors possess several characteristics that differ from commercially available bench-scale calorimeters reactors, such as using water as a cooling fluid and a relatively high residence time of the cooling fluid in the jacket. Moreover, the temperature control system is typically less efficient

compared to commercialized calorimeters, which can cause undesirable temperature oscillations. The application of calorimetric techniques in a standard laboratory reactor allows greater use of the calorimetric technique in research centers, development of industries, and academy. This is beneficial as commercial bench-scale calorimeter reactors have a very high cost, and their application is restricted to large research centers (Esposito; Sayer & Araújo, 2010; Rincón, *et al.*, 2013).

The present work features a standard laboratory reactor comprising a jacketed stainless-steel tank with an internal volume of 5 liters instrumented to operate as a calorimetric reactor. An algorithm was developed and implemented for measuring the cooling fluid flow rate and a methodology capable of identifying the loss of stability of the latex. The AquiDtool Calorimetry, a custom-developed supervisory system capable of performing the reactor temperature control and estimating monomer conversion in real-time (tool for data acquisition and calorimetry), allowed defining a temperature set point or following a reference trajectory. Two highly significant aspects regarding temperature control were analyzed. One of them is the effect of cooling fluid flow rate, which is related to the amount of heat that can be withdrawn directly through the jacket. The other relates to the presence of disturbances, for instance, in situations in which temperatures do not follow a specific dynamic behavior.

Real data provided an estimation for modeling the process and thus originated the transfer function of the controllers. In the case of the PID (Proportional, Integral, and Derivative (Seborg; Edgar & Mellichamp, 1989; Djarum & Ahmad, 2020) structure type derivative action was ignored (set equal to zero) because the derivative action tries to fix the oscillatory state (increasing the damping coefficient) which strongly demands the actuation of automatic valves diminishing its useful life (Ni; Debelak & Hunkeler, 1997). The IMC (Internal Model Control) (Brosilow & Joseph, 2002; Camacho & Bordons, 2004; Wang, *et al*., 2020) structure type ensured the system stability even in extreme cases, such as the variation of the cooling fluid flow rate and change in both operating mode and feeding of reactants with temperatures very different from the reaction medium. Both structures were insensitive to small physical

transitions resulting from modifications in the positioning and calibration of temperature sensors.

2 Material and methods

2.1 Experimental Setup

The reactor setup includes a data acquisition board that enables remote monitoring of the temperature system. Temperature data were acquired by four platinum resistance thermometers (PT100, class A) positioned at different locations in the reactor (jacket inlet and outlet, reaction medium, and surroundings). The heating/cooling fluid flow rate was continuously acquired throughout the reactions by a turbine single-jet extra dry water meter installed at the jacket outlet. The cooling fluid flow rate varied among the different reactions with a valve installed at the entry of the jacket.

A PI (Proportional and Integral) controller or an IMC (Internal Model Control) controlled the temperature within the reactor by synchronizing two pneumatic valves. These valves regulate the steam dosage (supplied by a boiler) via the transfer of heated and cooled water through the jacket.

The centrifugal pump operation initiated the heat exchange system, thereby maintaining constant pressure in the jacket. The steam valve was responsible for the steam circulation in the heat exchanger, which generated a warm current for heating the reactor. With the cold water valve closed, only warm current circulates in the reactor, receiving more energy in each passage through the heat exchanger. In this way, it reaches high temperatures in a relatively short time interval. The opening of the cold water valve decreases the pressure in the jacket and the line of circulation and forms a cold current. The cold water valve and the steam valve are proportional valves that allow to obtain the reaction temperature. A gas-heating boiler provided steam. Due to its control system, the boiler supplies steam at pressures ranging from 6 to 8kgf/cm^2 characterizing a source of disturbance to the polymerization process. The cooling tower supplied cold water, which was previously at ambient temperature. A centrifugal pump guaranteed the cooling's fluid circulation under

constant pressure between the tower and the single-flow valve installed in the cold water inlet system.

All equipment used in the plant, except for the pilot reactor, is meant for industrial use, which means that the accuracy of each piece of equipment is limited. Oscillations in the laboratory electric network, in the compressed air line feeding the control valves, and especially in the steam line, interfere directly with the quality of the sampled signals and the quality of the latex produced. In addition, the plant is multipurpose for not being used only for calorimetric studies.

2.2 Proportional, Integral and Derivative Control Structure Type

The proportional, integral, and derivative (PID) control strategy type was performed, after process model identification, according to the method of Sundaresan and Krishnaswamy (Seborg, *et al*., 1989). The method proposed by Cohen and Coon was used to perform the proportional integral (PI) controller's tuning (Seborg, *et al*., 1989). The derivative part of the PID controller was set equal to zero. It occurs because the derivative action tries to fix the oscillatory state (which increases the damping coefficient), demanding strong actuation of automatic valves diminishing their usability (Ni; Debelak & Hunkeler, 1997).

2.2.1 Sundaresan and Krishnaswamy's Method

This method avoids the inflection point construction's use entirely to estimate the time delay. The authors proposed that two times (t_1 and t_2) were estimated from a step response curve, corresponding to 35.3 and 85.3% response times, respectively. The time delay (θ) and time constant (/) are then estimated from Equation 1 e 2 (Seborg, *et al*., 1989, Roslan; Abd Karim & Hamzah, 2018).

$$\theta = 1.3\, t_1 - 0.29\, t_2 \quad (1)$$

$$\tau = 0.67\left(t_2 - t_1 \right) \quad (2)$$

These values of ⁄ and θ approximately minimize the difference between the measured response and the model response based on a correlation for many data sets.

2.2.2 Cohen and Coon Method

This controller design relations are based on a specific model named the first-order plus time-delay model. Cohen and Coon reported design relations developed empirically to provide closed-loop responses with a decay ratio of ¼, which refers to the ratio between two successive peaks of a damped oscillation.

2.2.3 Velocity Algorithm for digital PID Control

The PID controller used, Equation 3 written in the velocity form is obtained from the discretization of the continuous form of the PID (Seborg, *et al*., 1989).

$$U_t = U_{t-1} + K_c \left[\left(e_t - e_{t-1} \right) + \left(\Delta t / \tau_I \right) e_t + \left(\tau_D / \Delta t \right) \left(e_t - 2\, e_{t-1} + e_{t-2} \right) \right] \quad (3)$$

where U_t is the controller output at the t_{th} sampling instant, Δt is the sampling period (the time between successive samples of the controlled variable), and e_t is the error at the t_{th} sampling instant. K_c : controller gain.

The split-range variable structure control performed the hot and cold currents manipulation. To manipulate steam currents and cold water, we can use two approaches, split-range and independent variables manipulation types. In split-range control (divided control), only one controller is required, as it determines the signal to one of the manipulated variables, usually the one with

direct action (in this case, the opening of the steam valve), as the other variable changes in the opposite direction and equal proportion. The manipulation of two variables with the split-range strategy is interesting as it requires less computational effort in the controller implementation. However, it is very restrictive as it forces contradictory actions in the intermediate control region which corresponds to simultaneous heating and cooling.

2.3 Control Structure Type Internal Model Control

The control structure types Internal Model Control (IMC) depends on a transfer function model of the process previously obtained through an identification method (Brosilow & Joseph, 2002, Mien, 2019, Khanduja & Bhushan, 2016, Kumar; Nagy & Varga, 2015). The employed controller has only one parameter to be changed in real-time, which facilitates the adjustment and implementation thereof. The said parameter, called ACCELERATOR, represents approximately the relationship between the response times of open-loop and closed-loop processes.

To determine the process model, cold water and steam valve openings were manipulated in the form of step signals. For example, given the reactor carried out with water with the stirring system running and operating in an open-loop at a predetermined temperature and flow rate in the jacket, if the desired steady state was reached at 60% opening steam valve and 40% of cold water valve and the stipulated step is 20%, the steam valve must pass to 40% and the cold water to 60% opening instantly. The temperature variation obtained from this operation served as the basis for determining the process model.

The adjustment of the process model was realized by varying the dead time value of the model, the analysis error histogram, and the squared correlation coefficient. Another point observed during the identification was the time constants values of the second-order models showed that both time constants must have values considerably higher than the value of the sampling time (10s). Otherwise, the model should be considered first-order. This action is related to the controller parameters calculation to be real numbers. Since the time constant value of the process was much higher than the dead time encountered during the identification of the process model, this was disregarded

in the model used by the controller. It simplified the design of controllers by avoiding the use of dead time compensators, such as the Smith predictor (Camacho & Bordons, 2004) structure type.

One of the purposes of this work was to evaluate the effect of cooling fluid flow rate in isothermal and isoperibolic conditions. Hence was formulated a combination of four distinct possibilities of reactor operation. In this context, the process model was identified at high and low fluid flow rates to increase the controller's robustness. Two models were thus established for each fluid flow, an isothermal and an isoperibolic model.

2.3.1 Development of Empirical Models from Process Data

In the present work, an algorithm produced in Matlab computational language determined and analyzed the model.

Statistical analysis can be used to estimate unknown model parameters and specify the uncertainty associated with the empirical model. It can also be used to compare several candidate models. For linear models, the least-square approach is used to estimate model parameters. The least-squares method is the standard approach for calculating the values of the model parameters to be estimated that minimize the sum of the squares of the errors *S* for an arbitrary number of data points, *N* (Seborg, *et al*., 1989).

The unknown parameters of the model are those which minimize the sum (Adamczevski, 2011):

$$S = \sum_{i=1}^{N} \left(y_i - \hat{y}_i \right)^2 \qquad (4)$$

In Equation 4 the value of y_i is known (obtained experimentally), while $\hat{y}_i$ should be calculated (as an output of the model) so as to minimize *S*, the objective function. The minimizing of the sum-of-squared residual error is optimal only when the measurement errors have Gaussian distributions. *y*: system output, $\hat{y}$: output of the system model.

The current techniques utilized to obtain models involve solving Equation 4 using data provided by the system when a known input is applied. This data can be represented in the time domain by instantaneous values that vary along it or data in the frequency domain by referring to magnitude and phase values.

A system subject to adding a noise or disturbance $\xi(k)$ modeled by a function $H(q)$ in the system output, can be represented by the mathematical expression given by (Adamczevski, 2011):

$$y(k) = G(q)\,u(k) + H(q)\,\xi(k) \tag{5}$$

Based on Equation 5, five models of parametric structures can be defined: Finite Impulse Response (FIR), autoregressive with exogenous input (ARX), autoregressive moving average with exogenous input (ARMAX), output-error (OE) and Box-Jenkins (BJ). Which differ as to the modeling both in the input signal $u(k)$ as in relation to noise $\xi(k)$.

$$A(q)\,y(k) = \frac{B(q)}{F(q)}\,u(k) + \frac{C(q)}{D(q)}\,\xi(k) \tag{6}$$

where $A(q) = 1 + a_1\,q^{-1} + a_2\,q^{-2} + \ldots + a_{n_a}\,q^{-n_a}$, $B(q) = b_1\,q^{-1} + b_2\,q^{-2} + \ldots + b_{n_b}\,q^{-n_b}$,

$C(q) = 1 + c_1\,q^{-1} + c_2\,q^{-2} + \ldots + c_{n_c}\,q^{-n_c}$, $D(q) = 1 + d_1\,q^{-1} + d_2\,q^{-2} + \ldots + d_{n_d}\,q^{-n_d}$,

$F(q) = 1 + f_1\,q^{-1} + f_2\,q^{-2} + \ldots + f_{nf}\,q^{-nf}$

And q represents the unit-time advancement operator. Selecting specific values for $A(q)$, $B(q)$, $C(q)$, $D(q)$, and $F(q)$ allows changing between the different structures that can be used for identifying a system. A, B, C, and D: matrices of a state space representation.

In this work, a structure called output-error (OE) was used. Following this structured method, prediction error compares the measured value with the predicted value for the output. This structure has as output a signal without any

disturbance combined with an error or signal disturbance. The structure output-error (OE) is a special case of the general model (Equation 6), in which polynomials $A(q)$, $C(q)$, and $D(q)$ have unitary values, resulting in:

$$y(k) = \frac{B(q)}{F(q)}\ u(k) + \xi(k) \tag{7}$$

The model was made using two Matlab functions, IDDATA and OE. These functions can be used in both time and frequency domains. Therefore in this work, they were used in the time domain. Before performing the identification of the process model, for proper use of the functions IDDATA and OE, the operation point of the variables involved was subtracted, which were the steam valve opening, the reactor temperature (T_r) in isothermal mode, and the jacket outlet temperature (T_{jout}) in isoperibolic mode.

IDDATA creates a data object to encapsulate the input/output data and their properties, i.e. it creates a data object with output *Y* and input *U* and sampling interval t_s = 1. The object created with the IDDATA is used as input for the OE function, along with the dead time values and the order of the desired model. OE estimates output-error polynomial model using time domain data. The estimated model is delivered as an IDPOLY object (polynomial model with identifiable parameters). The identified output-error polynomial model contains the estimated values for *B* and *F* polynomials along with their covariances and structure information.

The idea of the quality of a model lies in the fact that it accurately represents the desired characteristics of a system and not necessarily all of them (Adamczevski, 2011). Two analyses were carried out for evaluating the error between the real and the estimated data: one graphical and the other using statistical tools. The FILTER function filters a data sequence using a digital filter which works for real and complex inputs. The output of the causal, linear, and time-invariant system for a given input when the system is specified by a linear differential equation with constant coefficients is shown next. *Ymodel*= FILTER (*B*(*q*), *F*(*q*), (steam valve (%) - operating point (%))) filters the

data in vector (steam valve (%) - operating point (%)) with the filter described by vectors *F* and *B* to create the filtered data *Ymodel* which equals to the vector (temperature (°*C*) - operating point (°*C*)). With the filtered and experimental data were calculated the standard deviation (STD) and the mean value of the error (MEAN) determined the squared correlation coefficient and generated a histogram of the error (HIST and NORMPDF).

2.3.2 Properties of IMC: Transfer Functions (Brosilow & Joseph, 2002).

1) Definition of the process transfer function. Analysis in discrete time using the *Z* transform:

$$P(z^{-1}) = \frac{b_0 z^{-1}}{1 + a_1 z^{-1} + a_2 z^{-2}} \tag{8}$$

2) Definition of the process transfer function in positive powers of *Z*:

$$P(z) = \frac{b_0 z}{z^2 + a_1 z + a_2} \tag{9}$$

3) Definition of the transfer function of IMC filter in *Z*:

$$Q(z) = \frac{z^2 + a_1 z + a_2}{b_0} \frac{(1 + t_1)^2}{(z + t_1)^2} \tag{10}$$

The first term of *Q(z)* is the inverse dynamics of the process (with no dead time). The second term is the filter with a unitary gain which makes the transfer function *Q(z)* physically realizable.

4) Product of *P(z)* and *Q(z)*:

$$Q(z)P(z) = \frac{b_0 z}{z^2 + a_1 z + a_2} \frac{z^2 + a_1 z + a_2}{b_0} \frac{(1+t_1)^2}{(z+t_1)^2} = \frac{(1+t_1)^2 z}{(z+t_1)^2} \tag{11}$$

5) Project of the controller *C(z)*:

$$C(z) = \frac{Q(z)}{1 - P(z)Q(z)} = \frac{\dfrac{z^2 + a_1 z + a_2}{b_0} \dfrac{(1+t_1)^2}{(z+t_1)^2}}{1 - \dfrac{(1+t_1)^2 z}{(z+t_1)^2}} = \frac{(1+t_1)^2}{b_0} \frac{z^2 + a_1 z + a_2}{(z+t_1)^2 - (1+t_1)^2 z}$$

$$C(z) = \frac{(1+t_1)^2}{b_0} \frac{z^2 + a_1 z + a_2}{(z+t_1)^2 - (1+t_1)^2 z} = \frac{(1+t_1)^2}{b_0} \frac{z^2 + a_1 z + a_2}{z^2 - (1+t_1^{\,2})z + t_1^{\,2}} \tag{12}$$

But:

$$C(z) = \frac{U(z)}{E(z)} = \frac{(1+t_1)^2}{b_0} \frac{z^2 + a_1 z + a_2}{z^2 - (1+t_1^{\,2})z + t_1^{\,2}}$$

$$\left[z^2 - (1+t_1^{\,2})z + t_1^{\,2} \right] U(z) = \frac{(1+t_1)^2}{b_0} \left[z^2 + a_1 z + a_2 \right] E(z) \tag{13}$$

6) Passing the controller to negative powers of *Z* (convenient for computer implementation) yields:

$$\left[1 - (1+t_1^{\,2})z^{-1} + t_1^{\,2} z^{-2} \right] U(z) = \frac{(1+t_1)^2}{b_0} \left[1 + a_1 z^{-1} + a_2 z^{-2} \right] E(z)$$

$$U(z) = \left[(1+t_1^{\,2})z^{-1} - t_1^{\,2} z^{-2} \right] U(z) + \frac{(1+t_1)^2}{b_0} \left[1 + a_1 z^{-1} + a_2 z^{-2} \right] E(z) \tag{14}$$

7) Controller in discrete time:

$$U(k) = \begin{bmatrix} cU1 & cU2 \end{bmatrix} \begin{bmatrix} U(k-1) \\ U(k-2) \end{bmatrix} + \begin{bmatrix} cE0 & cE1 & cE2 \end{bmatrix} \begin{bmatrix} E(k) \\ E(k-1) \\ E(k-2) \end{bmatrix}$$

$$U(k) = \begin{bmatrix} (1+t_1^{\,2}) & -t_1^{\,2} \end{bmatrix} \begin{bmatrix} U(k-1) \\ U(k-2) \end{bmatrix} + \frac{(1+t_1)^2}{b_0} \begin{bmatrix} 1 & a_1 & a_2 \end{bmatrix} \begin{bmatrix} E(k) \\ E(k-1) \\ E(k-2) \end{bmatrix} \tag{15}$$

2.4 AquiDtool CaloriMetry

The reaction temperature taken during the heating phase of reactants serves as a basis for determining the global heat exchange coefficient between the reaction medium and the jacket. For this reason, an algorithm capable of following trajectories defined by the user was developed. Initially, a matrix was set up. In the first matrix row are the initial temperature and time conditions .On the second row should be the temperature and the time required to reach such temperature As the interpolation occurs between two consecutive points, the time interval can be of any value that meets the desired trajectory. If, during the reaction, the time to achieve the trajectory is exceeded, the application AquiDtool Calorimetry will keep for an undetermined time the last temperature value read.

There are many possibilities for the operation and plant control which can be changed in real-time using AquiDtool CaloriMetry. In open-loop operation, it is possible to define the opening of the valves manually or opt for immediate cooling of the reactor, with 0% of the steam valve and 100 % of the cold water valve. In closed-loop operation, the setpoint and the ACCELERATOR values can be defined as isothermal calorimetry and/or isoperibolic calorimetry. Furthermore, this kind of control provides a heating curve. Another option in closed-loop is to use a reference trajectory previously defined. The trajectory measurement device (timer) allows the trajectory measurement initiates from *t=0s* once the application has already started. After some tests, the results showed that implementing the heating phase using step signals yields better results when operating in isoperibolic mode. As for the heating ramp, it works better while in isothermal mode.

2.5 Materials

The reactions utilized monomers, namely vinyl acetate (VAc), styrene (Sty), and methacrylic acid (MAA), of analytical grade. Potassium persulfate P.A. ($K_2S_2O_8$), sodium bicarbonate ($NaHCO_3$), and sodium lauryl sulfate (SLS) were used, respectively, as initiator, buffer agent, and surfactant. All reagents were used as received without further purification. Distilled water served as the continuous phase.

2.6 Experimental Procedure

In reactions R1 to R6, the reactor was initially charged with water, buffer agent (sodium bicarbonate), and surfactant (sodium lauryl sulfate). Proceeding according to the following sequence stirring frequency was set to 400 *rpm* under a nitrogen atmosphere, and the reactor was heated to the reaction temperature set at 50 °*C*. After that, a monomer (vinyl acetate) was added to the reactor. When the reaction temperature was reached, again after the addition of the monomer and application of constant power (*P* = 200 *W*) by an electrical heater (determining the initial value of the overall heat transfer coefficient between reactor and jacket), the initiator was charged (aqueous solution of potassium persulfate pre-heated at a temperature of 40 °*C* just before feeding to the reactor) starting the reaction. The given reactions utilized the formulations presented in Table 1.

Table 1 - Formulation of emulsion polymerization reactions (R1 to R9).

Reaction		**VAc (*g*)**	**Sty (*g*)**	**MAA (*g*)**	**H_2O (*g*)**	**$NaHCO_3$ (*g*)**	**SLS (*g*)**	**$K_2S_2O_8$ (*g*)**
R1 - R6, R8	Initial Charge	1260			2940	8	10	9
R7		1134			2648	8	10	8
R9			1100	55	2510	2	14.5	4

R7		325			100	1	0.5
	Feeding						
R9			1100	55			

VAc: vinyl acetate, Sty: styrene, MAA: methacrylic acid, H_2O: water, $NaHCO_3$: sodium bicarbonate, SLS: sodium lauryl sulfate, $K_2S_2O_8$: potassium persulfate P.A.
Source: From the authors.

The semi-batch reactions commenced as conventional batch emulsion polymerizations. In the R7 reaction, the intermittent feeding of reactants began after 58 minutes of reaction time. In the R9 reaction, the reactant feeding began after 34 minutes of reaction time, with a flow rate feed of 0.5 g/s lasting for 38 minutes to enable semi-batch operation.

In all polymerizations, the reactants were pre-heated before being added to the reactor. In reactions R3, R4, R5, and R9, a low flow rate was used in the reactor jacket, whereas in all other reactions high flow rates were used (Table 2). In order to validate conversion, estimated online samples were collected during the reaction for gravimetric analysis. These samples were put into preweighted aluminum capsules that already contained around 0.2 g of 1wt.-% hydroquinone aqueous solution (the exact amount was measured for each sample). Capsules were dried at 60 *ºC* until a constant weight was obtained.

Table 2 - Differences among emulsion polymerizations.

Reaction	**Heat-up phase**	***T*** **(*ºC*)**	**Solids content (*wt.-%*)**	**Method of reaction calorimetry**	**Average jacket flow rate (*L/min*)**	**Batch**	**Semi-batch**
R1	Curve	50	30	Isothermal	21	Yes	
R2	Curve	50	30	Isothermal	21	Yes	
R3	Curve	50	30	Isothermal	4	Yes	
R4	Curve	50	30	Isothermal	4	Yes	
R5	Curve	50	30	Isothermal / Isoperibolic	4	Yes	
R6	Curve	50	30	Isothermal / Isoperibolic	22	Yes	
R7	Ramp	50	35	Isothermal	22		Yes
R8	Steps	50	30	Isoperibolic	20	Yes	

R9	Steps	70	48	Isoperibolic	4	Yes

Source: From the authors.

3 Results and Discussion

Table 1 shows the reagents used in each reaction, the initial mass charge, and the mass feed. Table 2 shows an overview of the carried-out reactions. By independently or collectively varying the reaction temperature, the jacket coolant flow rate, and the solids content, different results can be obtained. This makes it an effective method for evaluating temperature control.

Development of empirical model from process data: Sundaresan and Krishnaswamy method parameters were determined based on Equation 1 e 2 (Seborg et al., 1989): $T_1 = 45.06\ °C$, $T_2 = 68.66\ °C$, $t_1 = 15.35\ min$, $t_2 = 52.20\ min$, $\theta = 4.82\ min$, $\tau = 24.69\ min$ and $K = 2.36\ °C/\%$. The PID controller tuning was based on Cohen and Coon method: $K_c = 2.44\ \%/°C$, $\tau_I = 571.80\ s$, $\tau_D = 87.67\ s$. And for the PI situation: $K_c = 1.5\ \%/°C$, $\tau_I = 500\ s$.

In reactions R2 and R4 (Figure 1), an inhibitor solution was added, corresponding to 10 mL of an aqueous solution containing 1% hydroquinone. The dashed line indicates the time when the inhibitor was added. The disturbance pointed out by number (1) shown in Figure 1 refers to monomer feeding. At this moment, the mass of the reagent undergoing heating from ambient temperature to the reaction temperature was approximately 2738 g. The mass of the monomer fed at 45 ºC, as indicated on the heating curve, was 1260 g. The monomer was left at ambient temperature.. The aim was to prevent monomer loss by evaporation, reducing its residence time within the same reactor. Although there has been a rise of approximately 15 ºC in the jacket temperature at a low flow rate (R3), we can see that the set point has been reached again within a given time interval. The same occurred by applying constant power of 200 W (number (2) in Figure 1) and after adding the inhibitor to the reaction. Therefore, it can be said that the actions taken by the PI controller used to control the reactor temperature (Tr) are satisfactory. The oscillation around the desired value was ± 2 ºC which may result from the

controller tuning. However, it was found that the small changes in parameters already set resulted in an offset existence.

Figure 1 - Reactor temperature (Tr) control using a PI controller with high cooling fluid flow rate through the jacket, (R1 and R2) and with low cooling fluid flow rate (R3 and R4). Heating of the reactants, monomer feed (1), calibration of the reactor by adding a constant power to the reaction medium (2), polymerization (3) and disturbance caused by the addition of inhibitor during the reaction (---).

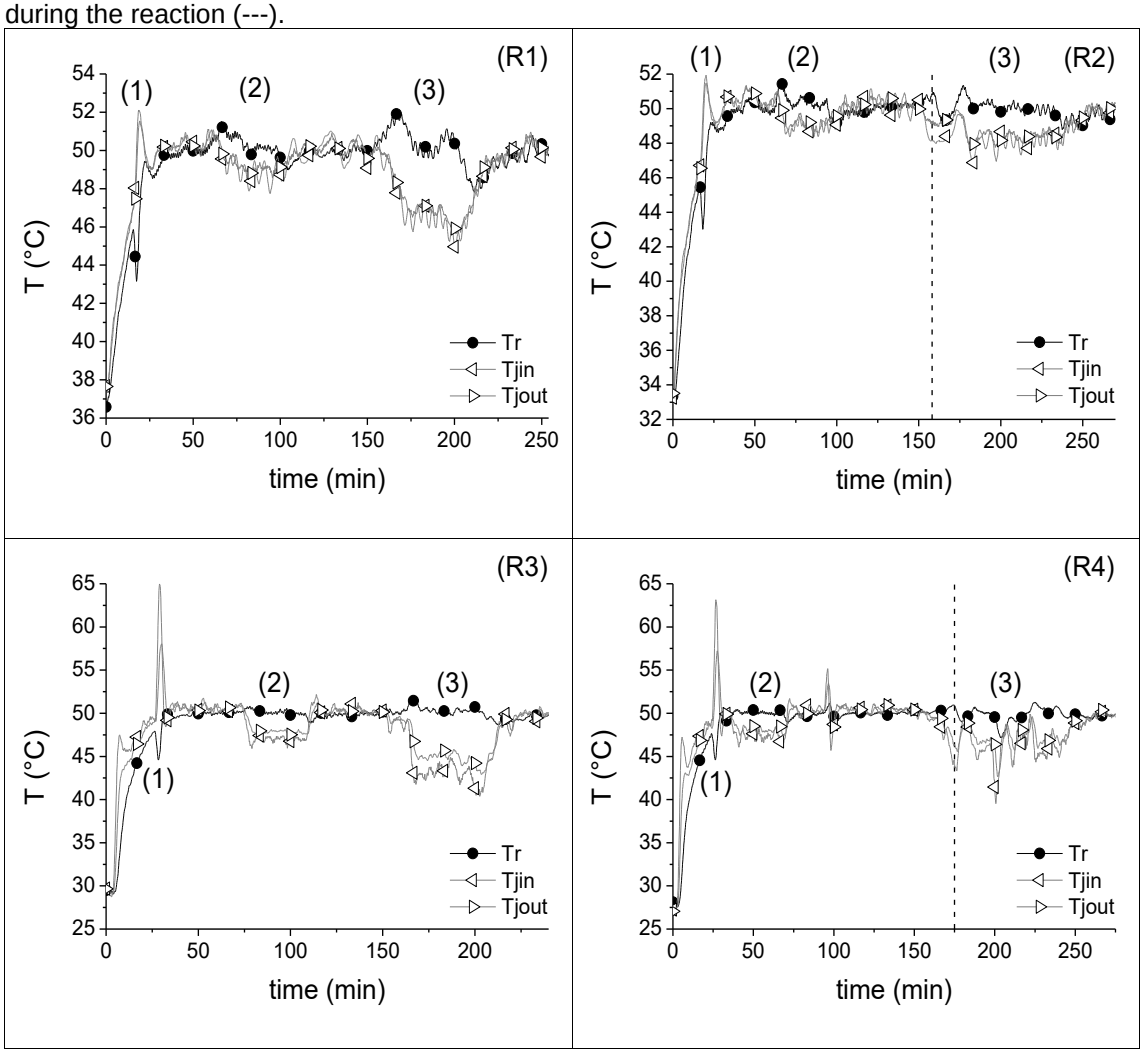


Figure 2 compares the model and real value Tr and Tjout with high and low cooling fluid flow rates through the jacket. During high flow rate conditions, the steam valve opening was decreased from 80% to 50%. Conversely, under low flow rate conditions, the reduction in valve opening was from 60% to 40% using split-range manipulation. These specific data points were used in identifying the process model. The opening percentage of the valves was chosen to obtain very different temperatures from the ambient temperature,

resulting in approximately 90 ºC at a high flow rate and around 80 ºC at a low cooling fluid flow rate. That is because the cooling tower operates at ambient temperature. The definition of which cooling curves would be used instead of heating the reactor comes from an experimental analysis of the process model identification, based primarily on the squared correlation coefficient. The data presented in Table 3 prove the good agreement between the experimental values and the process model. The existence of only a one-time constant (τ_1) makes it evident that the model in isoperibolic mode (T_{jout}) has lower order than the model in isothermal mode (T_r). It is also noticed that the time constant (τ_1) at a low flow rate has twice the value of the same constant for high a flow rate, unlike what happens with open-loop gain (K). The time delay (θ) in all cases has a value very close to the sampling time (10 s).

Figure 2 - Comparison between the model value and real value, a) and b) model Tr and Tjout, respectively, with high cooling fluid flow rate through the jacket and c) and d) model Tr and Tjout, respectively, with low cooling fluid flow rate through the jacket.

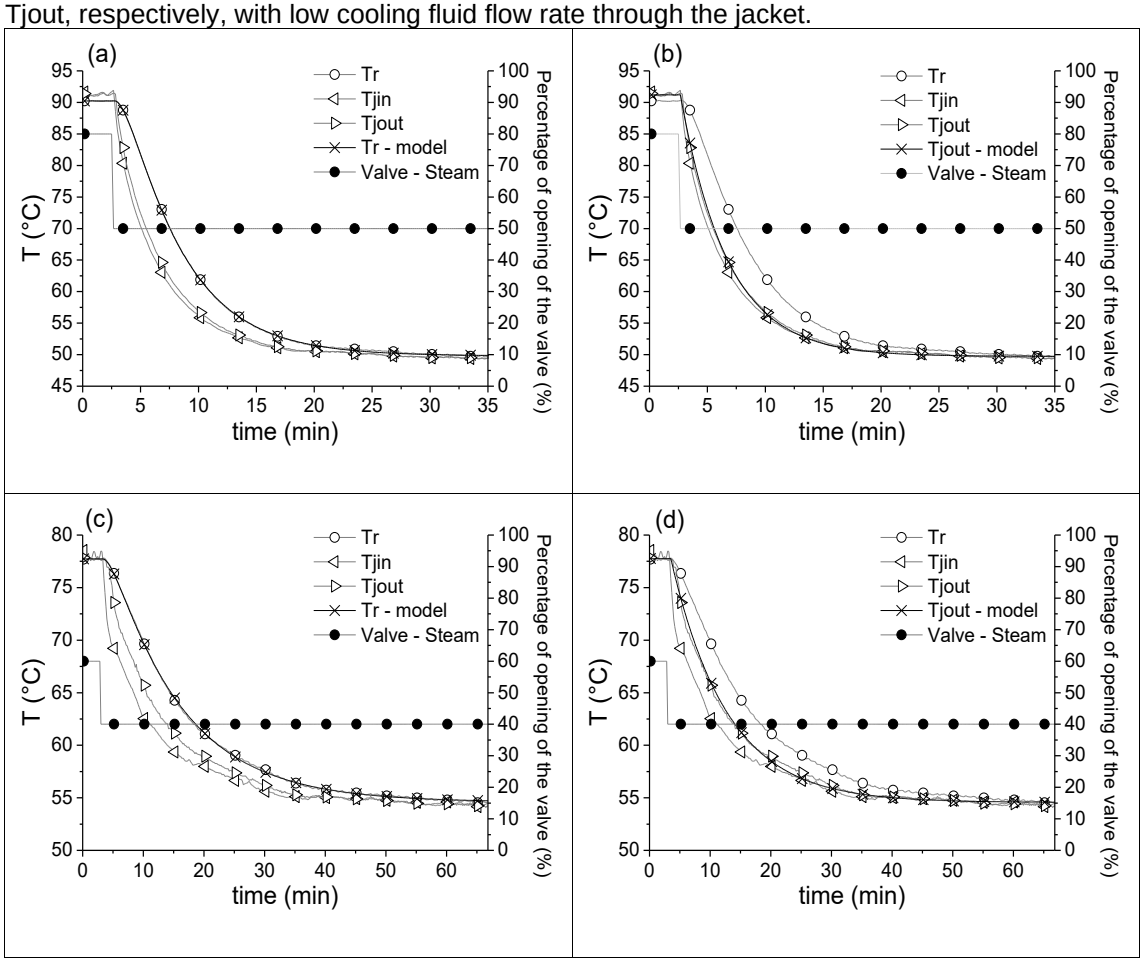


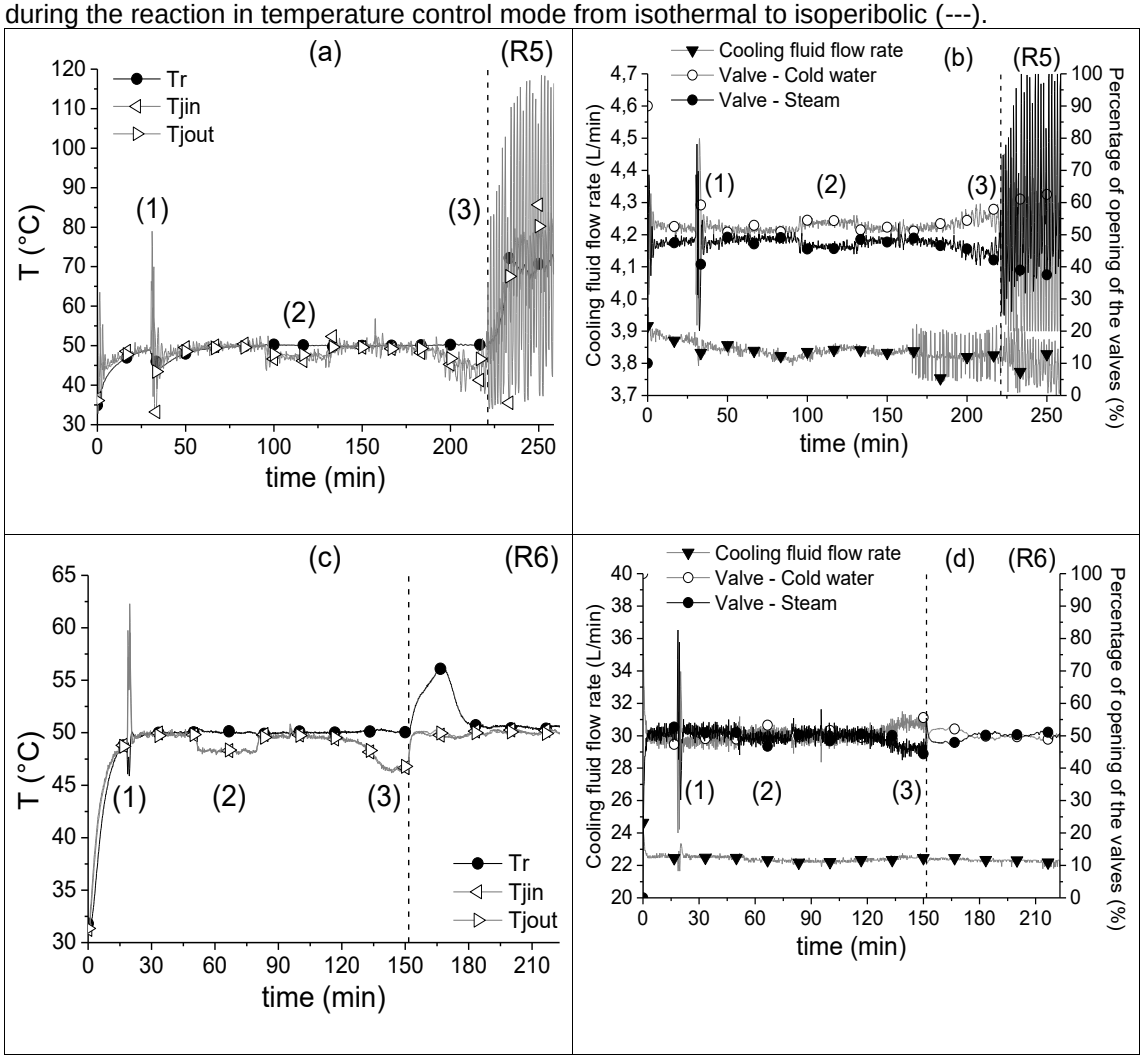
Table 3 - Parameters of the process model

Jacket flow rate / Model	***K***	***τ₁* (*s*)**	***τ₂* (*s*)**	***θ* (*s*)**	***r²* (%)**	**Discrete-time model:** $y(t)=[B(q)/F(q)]u(t)+e(t)$
High/T_r	1.35	300.27	72.15	10	99.99	$B(q)=5.716\times10^{-3}q^{-2}$
						$F(q)=1-1.838q^{-1}+0.842q^{-2}$
High/T_{jout}	1.38	247.33		0	99.92	$B(q)=5.48\times10^{-2}q^{-1}$
						$F(q)=1-9.604\times10^{-1}q^{-1}$
Low/T_r	1.16	718.16	98.29	20	99.98	$B(q)=1.548\times10^{-3}q^{-3}$
						$F(q)=1-1.889q^{-1}+0.891q^{-2}$
Low/T_{jout}	1.16	558.68		30	99.76	$B(q)=2.063\times10^{-2}q^{-4}$
						$F(q)=1-0.982q^{-1}$

K: gain, τ : controller time constant, *θ:* time delay***, r² s***quared correlation coefficient.
Source: From the authors.

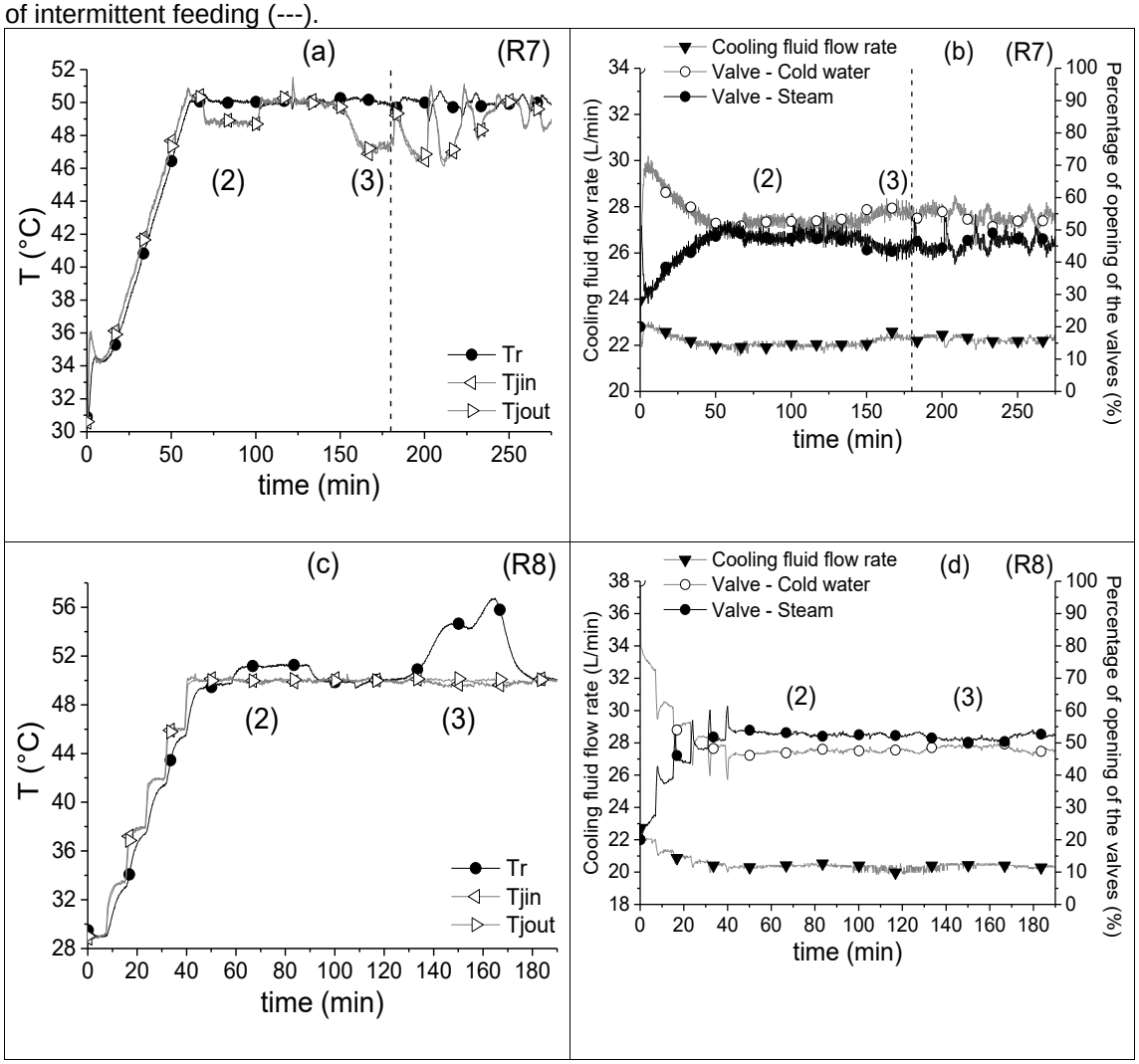
Referring to Figure 3 (c) R6 the excellent performance obtained can be confirmed after executing the control of the reaction temperature R6 since the oscillation around the set point was only of ± 0,5 ºC, either under isothermal conditions where T_r is the controlled variable or in isoperibolic conditions where T_{cs} is the controlled variable. On the R6, reaction the high flow rate of cooling fluid and the ACCELERATOR value reduction from 8 to 4 at the moment of the transition favored the temperature control. In the figure referring to the valve opening (Figure 3 (d) R6), it can be noticed that the control actions in the isoperibolic mode were smoother compared to the ones taken in isothermal the mode. This occurred due to the reduction of the ACCELERATOR value and because in isoperibolic conditions keeping only the temperature constant at the jacket outlet is sufficient, which reduces the order of the process model used by the controller from second to first order in both high and low flow rate.

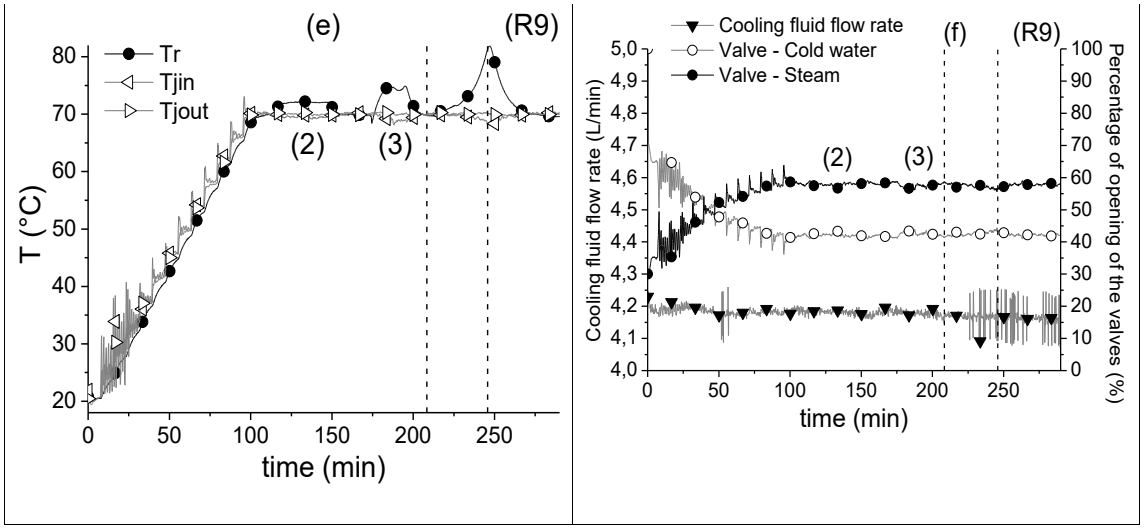
Figure 3 - Temperature control using a IMC controller with low cooling fluid flow rate through the jacket (a) R5 and with high cooling fluid flow rate (c) R6. (b) and (d) behavior of cooling fluid flow rate through the jacket and of heating and cooling fluid control valves during reactions R5 and R6, respectively. Heating of the reactants, monomer feed (1), calibration of the reactor by adding a constant power to the reaction medium (2), polymerization (3) and abrupt change during the reaction in temperature control mode from isothermal to isoperibolic (---).



The initiation of reaction R9 took place like a batch with relatively high final solid content. Using the conversion between 30% and 50% the intermittent feeding of the reactor was performed. In Figure 4 R9, the first dashed line indicates when feeding has started. The second dashed line indicates the end of the period. Figure 4 (f) R9 shows that the flow rate in the jacket remained stable, similar to the stability achieved in reactions involving VAc.

Figure 4 - Temperature control using a IMC controller with high cooling fluid flow rate through the jacket (a) R7 and (c) R8 and with low cooling fluid flow rate (e) R9. (b), (d) and (f) behavior of cooling fluid flow rate through the jacket and of heating and cooling fluid control valves during reactions R7, R8 and R9, respectively. Heating of reactants employing temperature ramp signal (a) R7 and employing temperature step signals (c) R8 and (f) R9, calibration of the reactor by adding a constant power to the reaction medium (2), polymerization (3) and start and end (R9) of intermittent feeding (---).





The effect of temperature reaction increase in comparison with the VAc polymerization can be identified by noticing a higher opening of the steam valve and a lower opening of the cold water valve. The ACCELERATOR value in reaction R9 was 4. This case shows that in isothermal mode, the variation from a high flow rate value of 8 to a low flow rate value of 2 is easily perceptible. On the other hand, in isoperibolic mode, the variation from a high flow rate value of 8 to a low flow rate value of 4 practically produces no difference in control actions.

4 FINAL CONSIDERATIONS

The temperature difference between the reactor and the jacket allows identifying the heat generated during the reaction. In most reactions presented, the heat peak was around 500 W which is a significant value once the reactor calibration with an electrical resistance with a power of 200 W was already represented as a load-type disturbance for the controller. Similarly, the initial monomer load feeding was also employed to assess the robustness of the PI and IMC controllers implemented. The performance of the controllers in these two cases was very similar and satisfactory. As there was no reaction in those cases, it was important that the system reached again, the set point in a time not too far and without saturation originating from the opening valves.

The disturbance caused by the presence of an inhibitor element did not strain the temperature control in the case of the PI controller. The same occurred with

the reactor's intermittent feeding while using the IMC controller. Real-time adjustment in reactor temperature control is particularly significant in situations that demand more effective actions. Such as with the heating of the reactants during the reaction when using reference trajectories as ramp or step signals and under abrupt transition from one operation mode to another. In these situations, only the IMC controller achieved good results because it takes into account a more accurate model of the process and mainly because of the "ACCELERATOR".

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