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Different Approaches in Concentration-Temperature Cascade Control of a Fixed Bed Reactor for the Phthalic Anhydride Synthesis

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This paper presents six different approaches for concentration-temperature type cascade control of a fixed bed reactor for the production of phthalic anhydride by oxidation of o-xylene. In the cascade scheme, a primary loop is responsible for controlling the phthalic anhydride exit concentration, while a secondary loop, based on temperature measurements in key positions along the reactor, maintains an optimal temperature profile. Simulations demonstrate the adequacy of the mean value of four temperature measurements monitored in the hot spot region to define the control action for manipulation of the heat rate that should be removed by the thermal fluid.

1. Introduction

Fixed bed reactors constitute complex systems used in several industrial chemical processes, and their operational control presents challenges, especially in the case of strongly exothermic reactions due to the effect of the inlet conditions on the temperature and concentration profiles along the catalytic bed. These reactors cannot be operated in excessive temperatures, since disturbances in inlet conditions can generate hot spots, with undesired consequences, such as thermal decontrolling, by-product formation from secondary reactions and catalyst deactivation (Karafyllis and Daoutidis, 2002).

According to Chou and Wu (2007), for fixed bed reactors where strongly exothermic reactions are carried out, it is necessary to control the temperature peak and exit concentrations simultaneously, to prevent excessive temperature elevations along the reactor and to guarantee the product specifications. Feasible scientific solutions have attracted the interest of lots of researchers for this question. Differently from the stirred tank reactors, fixed bed reactors are distributed systems, and maintenance of a prescribed thermal situation demands the adjustment of the temperature profile along the reactor, which requires the monitoring of this variable in some key positions.

In practice, due to physical restrictions and economic considerations, it is necessary to limit the number of sensors. Then, an important question in the controller design is the choice of the number and the optimal location of the temperature measurement points to obtain a good performance. The position and magnitude of the hot spot temperature cannot be established directly by non-linear explicit function, which imposes the use of feedback control strategies or steady state optimization (Wu and Chen, 2007).

In several industrial processes, it is impossible to measure the controlled variable with sufficient speed. This is the case, for example, for the concentration of chemical species in the exit of a fixed bed reactor. The measurement of this variable in real time is relatively expensive, and demands periodic calibration and maintenance of analyzers. Moreover, sampling rates are relatively slow due to the time required to purge the line connected to the reactor and the necessary time to the chromatography analysis (Budman et al., 1992).

Some strategies have been proposed for controlling fixed bed reactors, in which the objective is generally to make the control of the reactor exit concentration or the reaction conversion jointly with the hot spot

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temperature control, to prevent undesired thermal and hazardous situations. These strategies comprise sophisticated techniques of advanced control and simpler schemes based on the classical proportional-integral-derivative compensation, which is largely applied to the industrial process control, due to the ease of project and implementation. The control of tubular reactors with highly exothermic reactions by using of temperature measurement in three symmetrical axial positions has been considered by Urrea et al. (2008) and Hernandez-Martinez *et al*. (2010) that proposed efficient PID control structures for regulation of the concentration in reactor exit.

In this work, six concentration-temperature cascade schemes with four temperature measurements in hot spot region are proposed to control a fixed bed reactor for the strongly exothermic reaction of o-xylene oxidation used as an important route in industrial phthalic anhydride synthesis.

2. Mathematical Model for the Fixed Bed Reactor

Phthalic anhydride is an important intermediary of chemical and pharmaceutical industries, with a large application in the manufacture of dye, varnish, pigment and resin. The usual chemical route for its synthesis is the o-xylene oxidation carried out in gas phase, using a multitubular fixed bed catalytic reactor with a thermal fluid to remove the high heat rate generated by the strongly exothermic reactions which occur on the catalytic particles. The fixed bed reactor for the phthalic anhydride synthesis has been considered as a reference system in studies of some advanced control strategies applied to chemical systems (Chen and Sun, 1991; Hua and Jutan, 2000; Wu and Huang, 2003; Chou and Wu, 2007).

In modeling the reactor, the following basic hypotheses were considered: (i) one-dimensional model; (ii) plug-flow flux without axial dispersion; (iii) flux with negligible pressure drop; (iv) presence of external resistances to the mass and heat transfer; (v) parallel flux of the reaction mixture and thermal fluid. Three basic reactions are considered, as the kinetic scheme proposed by Froment (1967): the desired main reaction of o-xylene oxidation to give the phthalic anhydride, the undesired consecutive reaction of phthalic anhydride to give carbon dioxide, and the parallel reaction of complete oxidation of o-xylene to give carbon dioxide. Jesus et al. (2010) show the complete modeling, numerical solution and parametric sensitivity study for an industrial operation condition considered for this process. Due to space limitation, the mathematical model is not presented in this paper.

3. Reactor Control

The proposed control problem consists of the regulation of the product concentration leaving the reactor through manipulation of the thermal fluid temperature. Variations of reactor inlet concentration and temperature of the reaction mixture are the disturbances that affect the process dynamic. As fixed bed reactors constitute distributed parameter system, in maintaining the desired thermal situation into bed it is necessary to monitor the temperature in some positions along the bed, mainly near the reactor inlet, where the hot spot occurs. For the control implementation were considered measurements of the temperatures, T_1, T_2, T_3 and T_4 in each sampling instant in the axial positions z_1 = 0,20 m, z_2 = 0,40 m, z_3 = 0,60 m and z_4 = 0,80 m, for L = 4 m (reactor length).

Cascade structures proposed in this work has one typical configuration, showed in Figure 1, being formed of two loops, one designed as primary or master loop for the concentration control and the other one designed as secondary or slave loop for the temperature control.

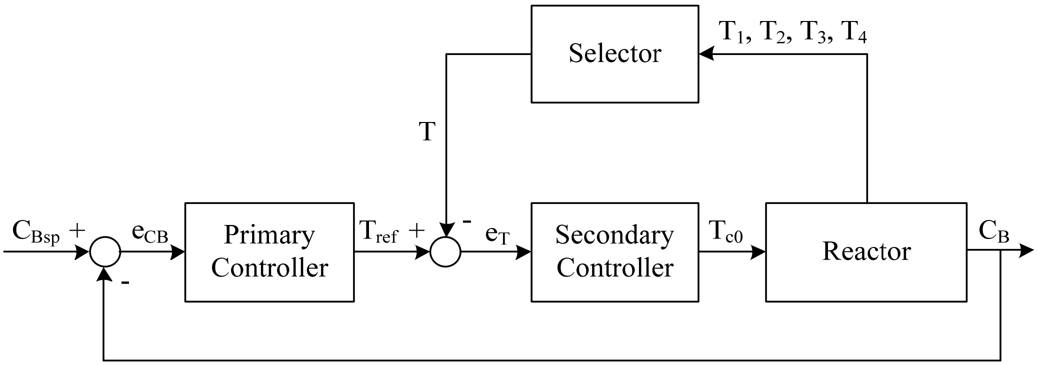

Figure 1: Schematic diagram of concentration-temperature cascade control for the fixed bed reactor.

Six concentration-temperature cascade schemes were designed, with measurements of the phthalic anhydride concentration in reactor exit and temperature in different positions along the reactor for each sampling instant, according to the proposed secondary loop configuration, namely: Cascade scheme 1 – Only the temperature T_1 is measured; Cascade scheme 2 – Only the temperature T_2 is measured; Cascade scheme 3 – Only the temperature T_3 is measured; Cascade scheme 4 – Only the temperature T_4 is measured; Cascade scheme 5 – The four temperatures T_1, T_2, T_3 and T_4 are measured and a mean value of these temperatures, T_m, is considered; Cascade scheme 6 – The four temperatures T_1, T_2, T_3 and T_4 are measured and the greater temperature is considered (selective control).

PI controllers compose each loop (primary and secondary) of the cascade schemes proposed and the control laws are given by the following relations:

$$\left(T_{c0}\right)_k = \left(T_{c0}\right)_{k-1} + K_{c2}\left[\left(1+\frac{T_s}{\tau_{i2}}\right)\left(e_T\right)_k - \left(e_T\right)_{k-1}\right] \tag{1}$$

$$\left(T_{ref}\right)_k = \left(T_{ref}\right)_{k-1} + K_{c1}\left[\left(1+\frac{T_s}{\tau_{i1}}\right)\left(e_{CB}\right)_k - \left(e_{CB}\right)_{k-1}\right] \tag{2}$$

Where: K_{c1} – proportional gain of primary controller; K_{c2} – proportional gain of secondary controller; τ_{i1} – integral time of primary controller; τ_{i2} – integral time of secondary controller; T_{ref} – reference temperature for the secondary controller; T_{c0} – inlet temperature of coolant fluid (manipulated variable); T_s – sampling time.

4. Results

Simulations for each cascade scheme were carried out by using the mathematical model developed by Jesus et al. (2010) for an industrial operational condition of the phthalic anhydride synthesis process. The dynamical problem is established by supposing disturbances in feed conditions (T_0 and C_{A0}), which affect the behavior of the system. The regulation is obtained by manipulation of coolant temperature in reactor inlet.

For the testing of the six control schemes, a computational modulus in *Matlab* has been implemented. A sample time of 10 s and a delay time of 360 s for the concentration measurement have been considered in the calculus. The feed referential condition is C_{A0} = 0.1811 mol m^{-3} (feed concentration of o-xylene), T_0 = 628 K (feed temperature of reactants) and T_{c0} = 628 K (feed temperature of coolant fluid). For these operational conditions one obtains a concentration of 0.1335 mol m^{-3} as set point for the phthalic anhydride concentration in reactor exit. Tuning of the controllers was obtained by a trial and error procedure, considering a disturbance of 4% in the input variable T_0 (the reference value was changed from 628 K to 653.12 K). The Integral of the Square Error (ISE) was considered as principal criterion in defining the best parametric condition. Table 1 presents parameter values obtained for the primary and secondary controllers in each cascade scheme.

Table 1: Controller parameters tuned for a step disturbance of 4% in T_0 (T_0 = 653.12 K).

Parameter	Cascade 1	Cascade 2	Cascade 3	Cascade 4	Cascade 5	Cascade 6
K_{c1} (K mol m^{-3})	5300	5000	6000	5800	268	*
τ_{i1} (s)	350	300	800	470	90	*
K_{c2}	0.075	0.070	0.065	0.065	4.80	*
τ_{i2} (s)	400	450	350	400	82	*
T_s	46.67	47.33	58.50	54.17	18.67	48.33
ISE (mol^2 m^{-6} min^2)	4.01×10^{-5}	5.22×10^{-5}	6.07×10^{-5}	6.24×10^{-5}	1.31×10^{-5}	2.72×10^{-5}
IAE (mol m^{-3} min)	0.0308	0.0369	0.0406	0.0435	0.0139	0.0267
max(T(t)) (K)	665.85	671.68	671.97	671.12	664.25	667.62

* For this controller, the parameter values of the cascade schemes 1, 2, 3 or 4 are assumed according to the position where the maximum temperature selected occurs in each instant of the regulation action.

Figure 2 shows responses of the controlled and manipulated variables for the tuning test, which has been implemented by a step disturbance in inlet temperature of the reactants. The cascade scheme 5, which considers the mean temperature of the four measurements, presents the best performance in terms of the Integral of Square Error (ISE) and the Integral of Absolute Error (IAE), as can be seen by the numerical values showed in Table 1. Additionally, the temperature peak, max (T(t)), attains the smaller value for this cascade control scheme (664.25 K).

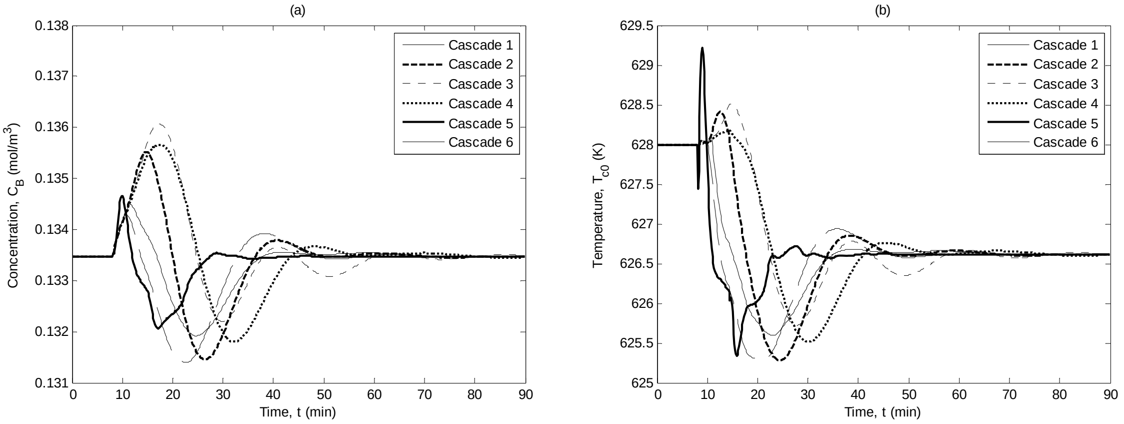

Figure 2: Response of controlled (a) and manipulated (b) variables in the tuning test.

With the tuned controllers, several test simulations have been implemented by successive disturbances in the reference values of T_0 and C_{A0}. Figure 3 shows the responses of the controlled and manipulated variables. One obtains the same conclusions of the tuning test, which demonstrate the excellent performance of the cascade scheme 5, presenting a faster stabilization of the phthalic anhydride concentration in reactor exit by manipulation in the inlet temperature of the coolant fluid used for heat removal from the catalytic bed.

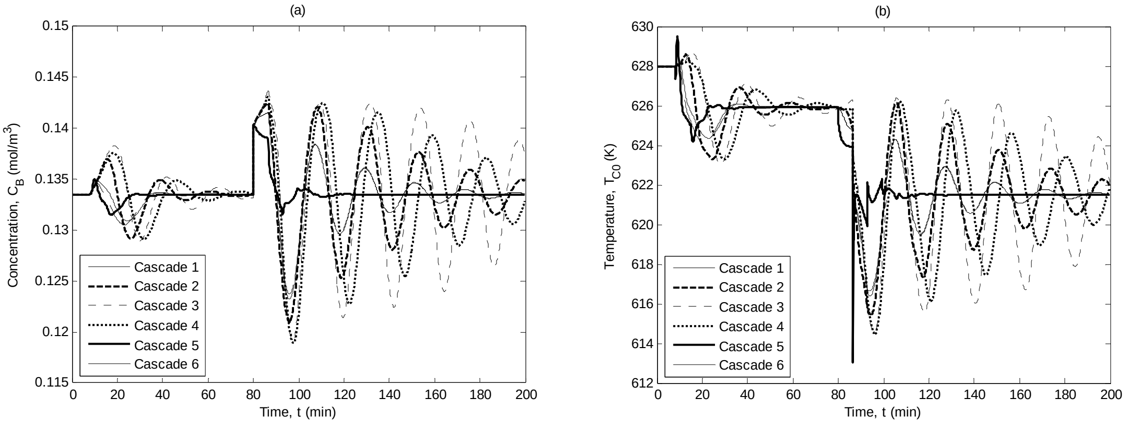

Figure 3: Response of controlled (a) and manipulated (b) variables for successive disturbances in T_0 and C_{A0}.

Figures 4 and 5 show the temperature profiles in each point of measurement for the test simulation by using the six proposed schemes of concentration-temperature cascade control for the fixed bed reactor. The very good performance of the cascade scheme 5 can be again highlighted, which exhibits an efficient regulation of the temperature profile along the catalytic bed, especially in the critical region of hot spot formation. This is fundamental to prevent the evolution of excessive thermal profile and to guarantee the product quality and safe operational condition.

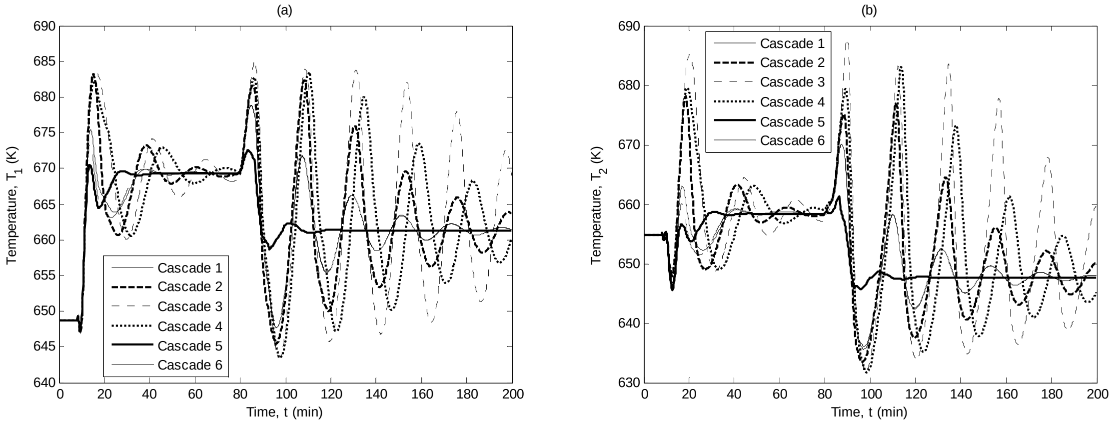

Figure 4: Temperature profiles in z = 0.20 m (a) and z = 0.40 m (b) for successive disturbances in T_0 and C_{A0}.

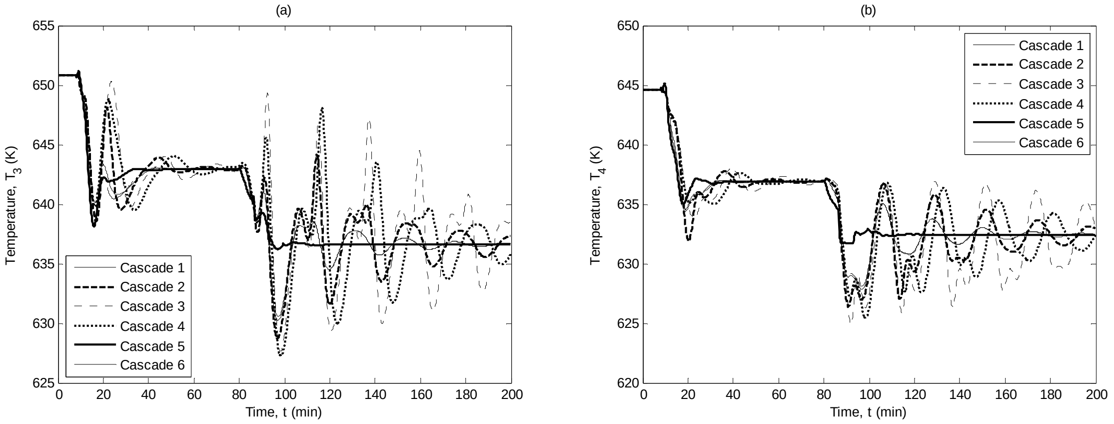

Figure 5: Temperature profiles in z = 0.60 m (a) and z = 0.80 m (b) for successive disturbances in T_0 and C_{A0}.

Finally, Figure 6 exhibits the accumulated profiles of Integral of Square Error (ISE) and Integral of Absolute Error (IAE) for the test simulation. The least values for these performance parameters are obtained for the cascade scheme 5 which uses the mean temperature of T_1, T_2, T_3 and T_4 to attain the reactor control objective.

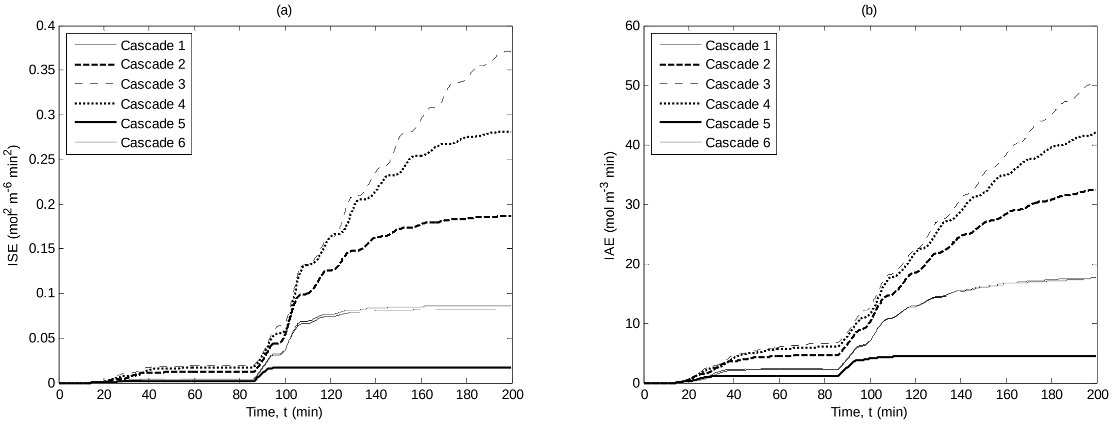

Figure 6: Accumulated profiles of ISE (a) and IAE (b) for successive disturbances in T_0 and C_{A0}.

5. Conclusions

Results of simulations for six proposed cascade control schemes showed the good performance of the scheme that considers the phthalic anhydride concentration in reactor exit and four temperature measurements in hot spot region (cascade scheme 5). In this scheme, a mean value of temperature, T_m, is employed for the temperature control by the secondary controller of the concentration-temperature cascade scheme. This strategy has a more efficient performance, than in the case where only a temperature measurement in a specific point is considered, or when it is considered the scheme of selective control usually employed in the control of fixed bed reactors. The strategy analyzed is similar to the methodology proposed by Urrea *et al.* (2008) and Hernandez-Martinez *et al.* (2010). However, these authors consider a weighted mean temperature, using arbitrary weights for each temperature measurement in different axial positions. The obtained results in this present work prove the possibility of designing efficient regulatory structures combining the well-established conventional control laws which are preferentially chosen in the industrial practice.

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