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MULTIVARIABLE CONTROL OF AN INDUSTRIAL GAS PHASE COPOLYMERIZATION REACTOR

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Abstract
Advanced control is used to avoid large temperature offsets for a batch copolymerization gas phase reactor used for test of catalysts. A nonlinear state-space model is developed to represent the reactor behavior in terms of mass and energy balances. A growth model describes the gas phase copolymerization and includes an activation-deactivation model for the catalyst. The multivariable control system includes three manipulated inputs, three controlled outputs and two measurements, temperature and pressure. The strategy combines nonlinear geometric control laws where states are either estimated by extended Kalman filtering or predicted from the kinetic model. This advanced control has been implemented with success on an industrial reactor.

Keywords: Multivariable control, nonlinear control, copolymerization, batch, reactor

1. INTRODUCTION

Polymerization is a major industrial domain where simulation, state and parameter estimation, advanced control can be very profitable due to the huge worldwide production. Among polymer products, polyethylene is the most common and has been the subject of many studies.

It is important to note that the model developed in the present paper for a batch reactor is very different from models that were developed for continuous fluidized bed reactors for which many articles are available. (McAuley *et al.*, 1990) devised a detailed kinetic model for industrial gas-phase ethylene and alpha-olefins copolymerization in presence of a multiple active site Ziegler-Natta catalyst. They thus predict the main properties of the polymer such as the copolymer composition and the molecular weight distribution assuming that the industrial reactor behaves as a CSTR where temperature is assumed to be well controlled and consequently uniform in the bed. (McAuley and MacGregor, 1993) were able to regulate the polyethylene melt index and density considered as averaged properties of the polymer and to follow grade changeovers by global input-output linearization which gave better results than linear IMC based on discrete transfer functions. States and qualities were estimated by extended Kalman filter and the grade changeovers were based on trajectories previously determined by offline open-loop dynamic optimization (McAuley and MacGregor, 1992). The nonlinear model used for control consisted of mass balances on hydrogen, butene, catalyst sites and polymer. The melt index and density were calculated from algebraic models. The manipulated inputs were the hydrogen and butene feed rates. In (McAuley *et al.*, 1995), the energy balance was considered and the stability issue already studied by (Choi and Ray, 1985) was addressed in the context of the possible addition of a gas recycle and a heat exchanger system to the model. Unstable behavior and limit cycles were thus obtained in absence of feedback control. (Dadebo *et al.*, 1997) also remarked a better performance of a nonlinear geometric controller compared to optimally-tuned PID. They studied the influence of different types of models of heat exchangers including staged ones. In studies by (Ali *et al.*, 1998; Ali *et al.*, 1999), only three states are necessary for optimization and PI or NLMPC control: the monomer and catalyst concentrations and the emulsion phase temperature. Their model allowed them to conduct a stability study where the catalyst injection rate and the ratio between the superficial velocity and the minimum fluidization velocity are the bifurcation parameters. They show that to operate safely with respect to polymer temperature, the polymerization reaction must be performed at low ethylene conversion unless good feedback control allows to maintain the temperature below the softening temperature of polymer to avoid agglomeration of the polymer particles.

In general, model predictive control is prefered as advanced control in industrial plants (Qin and Badgwell, 1996; Camacho and Bordons, 1995) due to its multivariable character and its capability to handle hard and soft constraints (Maciejowski, 2002). It may show limitations when faced to large nonlinearities, however nonlinear model predictive control (Allgöwer and Zheng, 2000) poses practical problems of implementation due to real-time optimization.

On the present copolymerization gas phase reactor working in batch mode, used to test highly reactive catalysts, undesired large temperature offsets were present with conventional PID control. Thus there was a possibility to demonstrate that advanced control could largely improve the reactor behavior. Nonlinear control (Isidori, 1995; Kravaris and Kantor, 1990*a*; Kravaris and Kantor, 1990*b*) was chosen because of its very reactive character.

2. MODEL OF THE REACTOR

This model is totally new and was built especially for two purposes. It should represent correctly the dynamic behavior of the copolymerization reactor with respect to temperature, pressure and chemical reactions without introducing unnecessary details about polymerization, such as the description of the distributions of molecular weights. Furthermore, its complexity should be in adequation with the future nonlinear control strategy to be used. The batch gas phase reactor is assumed to be well stirred by means of inert solids present during all the experiment. The model based on mass and energy balances takes into account eight states, respectively: temperature of solids and polymer, gas temperature, temperature of jacket fluid, pressure, monomer 1 and 2 concentrations, polymer concentration, particle radius.

$$\begin{aligned}\dot{x}_1 = &\frac{1}{M_{is}C_{pis} + M_pC_{pp}}\,[(-\mathcal{R}_1\Delta H_1 - \mathcal{R}_2\Delta H_2)V_g \\ &+U_{rj}\,A_{rj}\,(x_3 - x_1) + h_{gisp}\,A_{isp}\,(x_2 - x_1)]\end{aligned} \tag{1}$$

$$\begin{aligned}\dot{x}_2 = &\frac{1}{M_gC_{pg}}[F_{mono1}\,C_{p1in}\,(T_{1in} - x_6) \\ &+F_{mono2}\,C_{p2in}\,(T_{2in} - x_6) \\ &+h_{gr}\,A_r\,(x_3 - x_2) + h_{gisp}\,A_{isp}\,(x_1 - x_2)]\end{aligned} \tag{2}$$

$$\begin{aligned}\dot{x}_3 = &\frac{1}{M_jC_{pj}}\,[U_{rj}\,A_{rj}\,(x_1 - x_3) + \\ &F_j\,C_{pj}\,(T_{jin} - x_3)]\end{aligned} \tag{3}$$

$$\dot{x}_4 = R\,x_1\,\left(\frac{\dot{x}_5}{M_{mono1}} + \frac{\dot{x}_6}{M_{mono2}}\right) + R\frac{n_g}{V_g}\,\dot{x}_2 \tag{4}$$

$$\dot{x}_5 = -\mathcal{R}_1 + \frac{F_{mono1}}{V_g} \tag{5}$$

$$\dot{x}_6 = -\mathcal{R}_2 + \frac{F_{mono2}}{V_g} \tag{6}$$

$$\dot{x}_7 = N_p\,\mathcal{R}_{pol}\,\frac{1}{V_{ref}} \tag{7}$$

$$\dot{x}_8 = \mathcal{R}_{pol}\frac{1}{4\pi x_8^2\rho_{pol}} \tag{8}$$

Algebraic equations are added including in particular a growth model for the polymer grains inspired from (Choi and Ray, 1985; Kosek *et al.*, 2001).

The respective rates of reaction concerning monomers are

$$\mathcal{R}_1 = k_{p1}\,C_{cat}\,x_5 \quad ; \quad \mathcal{R}_2 = k_{p2}\,C_{cat}\,x_6 \tag{9}$$

and the rate of polymerization expressed for one particle is

$$\mathcal{R}_{pol} = \frac{(\mathcal{R}_1 + \mathcal{R}_2)V_g}{N_p} \tag{10}$$

with: N_p number of catalyst particles.

Catalyst concentration C_{cat} is expressed according to (Kosek *et al.*, 2001)

$$C_{cat} = \left(\frac{r_{part.cat.}}{x_8}\right)^3 \rho_{cat} \tag{11}$$

The kinetic constant for monomer i is (Kosek *et al.*, 2001)

$$k_{pi0} = \frac{yield}{M_{w,i}\,\exp(-\dfrac{E_{ai}}{Rx_1})\,C_i} \tag{12}$$

where the yield is expressed in kg polymer/kg catalyst/s.

The catalyst activity is inspired by an activation-deactivation model (Kittilsen *et al.*, 2001) which introduces empirical considerations to take into account the deactivation of the catalyst by modification of the propagation constant

$$\mathcal{R}_{pol} = \mathcal{R}_{pol}\;(1 - \exp(\frac{t}{t_{peak}}))\;\exp(-k_{deact}t) \tag{13}$$

and adapted to our case. t_{peak} is a parameter which controls the time to reach peak activity and k_{deact} is the deactivation constant (inverse of a time constant).

Open-loop experiments were performed on the reactor to identify the heat transfer coefficients.

3. CONTROL LAWS

3.1 Control strategy

Control variables are given in Table 1.

Table 1. Manipulated inputs and controlled outputs.

Manipulated input u_i	Controlled output y_i
Cold heat exchanger valve position u_1	Reactor gas temperature y_1
Mass flow rate of monomer 1 u_2	Pressure y_2
Mass flow rate of monomer 2 u_3	Ratio of monomer concentrations y_3

The measured reactor temperature can be adjusted by manipulating the inlet temperature of the heat-exchange fluid which circulates at constant flow rate split between two hot and cold heat exchangers similarly to (Corriou, 2004) according to Figure 1.

$$T_{jin} = u_1\,T_{cold} + (1 - u_1)\,T_{hot} \tag{14}$$

u_1 varies in $[0, 1]$. The mass flow rates of monomers influence the measured pressure, however monomer concentrations are measured with a large delay and inaccuracy and thus the system has only two available measurements. Consequently, the system is controllable but the ratio of monomer concentrations is not observable.

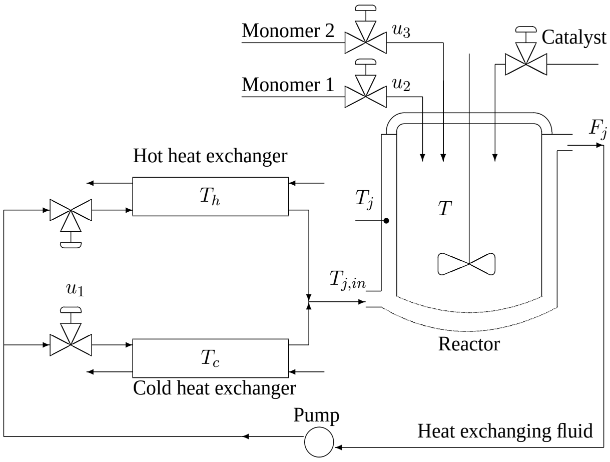

Figure 1. Experimental setup.

To avoid coupling problems with pressure due to the ideal gas law, the temperature control is considered as a SISO system. On the other hand, a multivariable controller is developed for the gas pressure and the ratio of monomer concentrations. To control this latter, the monomer concentrations are predicted from the kinetic model. Thus the only possible strategy is to impose a given ratio between the monomer flow rates in order to get the desired pressure and ratio of monomer concentrations.

The automation of the batch reactor successively includes reaching the temperature set point, the pressure set point, introducing the catalyst and starting the reaction. As the chemical reaction is likely to differently consume both monomers, different strategies must be used before and after start of reaction.

The end-user first requires that the peak of exothermal temperature occurring soon after the introduction of a very reactive catalyst does not exceed 5K. When the exotherm becomes dampened, the temperature should come back smoothly to its set point. Another requirement is that the pressure follows rapidly its set point. The delayed recording of monomers concentrations can show a posteriori if their ratio is respected by use of the control laws.

For temperature control, a SISO non linear control law (Isidori, 1995) is used. For pressure and ratio of monomer concentrations control, a MIMO non linear control law is used. An extended Kalman filter is used (Corriou, 2004; Grewal and Andrews,

2001) to estimate four of the eight states, i.e. the three temperatures and the pressure, based on the measurements of the gas-solids temperature and pressure. In absence of measurement of the monomer concentrations, these latter, the polymer concentration and the polymer particle radius are predicted from the nonlinear reactor model. The final control strategy is summarized in Figure 2. Due to the fast exothermicity of the reaction, a sampling period equal to $T_s = 2$s is chosen. The actual reactor presents a delay of about 40s of its temperature with respect to a move on the heat exchangers valves.

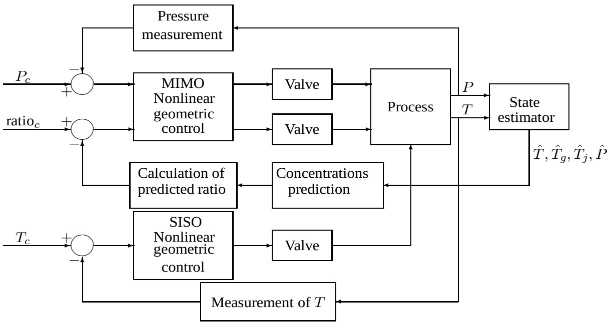

Figure 2. Control scheme.

3.2 Nonlinear control laws

The nonlinear control laws (Isidori, 1995; Corriou, 2004) are based on the theoretical model represented by the set of ordinary differential and algebraic equations. The nonlinear model of the reactor, affine with respect to the inputs, can be written in state-space form as

$$\begin{aligned} \dot{\boldsymbol{x}} &= \boldsymbol{f}(\boldsymbol{x}) + \sum_{i=1}^{3} \boldsymbol{g}_i(\boldsymbol{x})\, u_i \\ \boldsymbol{y} &= \boldsymbol{h}(\boldsymbol{x}) \end{aligned} \tag{15}$$

Presently, the relative order r is equal to 2 with respect to temperature and to 1 with respect to pressure. The nonlinear temperature control law is

$$u_1 = \frac{v_1 - \beta_2 \mathcal{L}_f h(\boldsymbol{x}) - \mathcal{L}_f^2 h(\boldsymbol{x})}{\mathcal{L}_g \mathcal{L}_f h(\boldsymbol{x})} \tag{16}$$

and the nonlinear pressure control law

$$u_2 = \frac{v_2 - \mathcal{L}_f h(\boldsymbol{x})}{\mathcal{L}_g h(\boldsymbol{x})} \tag{17}$$

where $\mathcal{L}_g h$ is the Lie derivative of h in direction of g. v is an external input to take into account modelling errors and disturbances. From calculated u_2, the positions of the two valves of monomers mass flow rates result. The tuning of the coefficients in both nonlinear control laws including the parameters of the external input is based on a pole placement.

States estimated by the extended Kalman filter (Corriou, 2004) and predicted states deduced from the estimated states are used in the control laws. In simulation, a Gaussian noise is added to the theoretical measurements.

The set points follow a sequence prescribed by the operator (Figures 3 and 8): after heating of the reactor, the monomers are introduced at a pressure slightly lower than the final pressure set point. Then the catalyst is introduced with nitrogen. Finally the chemical reaction starts at the required pressure. A ratio between the mass concentrations of monomers is imposed during the whole process. All the procedure was automated except for the introduction of the catalyst.

3.3 Results

For confidentiality reasons, the scales of the figures have been modified, however the deviations of the ordinate axis for the state variables are exact and the values of the manipulated variables have not been modified. Two parts are exposed: simulation and experimental results.

3.3.1. Simulation In simulation, a very reactive catalyst was chosen to demonstrate the potential of control. At initial time, the reactor temperature must reach its set point (Figure 3). During most of this stage, the valve of the cold heat exchanger is closed (Figure 4). The valve saturates in open position soon after $t = 2000$s when the catalyst is introduced into the reactor and provokes a large release of heat. It remains open until the catalyst activity decreases sufficiently.

The first pressure set point is imposed at $t = 800$s. The reactor pressure (Figure 5) follows very well the filtered set point. When the reaction occurs, first a deviation is seen between the actual pressure and its set point due to the coupling through the gas equation of state, then the pressure reaches its set point. In this case, a more reactive tuning would be needed for the actual reactor. The ratio of monomer concentrations is always perfectly followed in simulation.

3.3.2. Experiments On the actual reactor, measurements, valves automation including PID control, safety actions are ensured by an Eurotherm 2000 system operated according to OPC standard

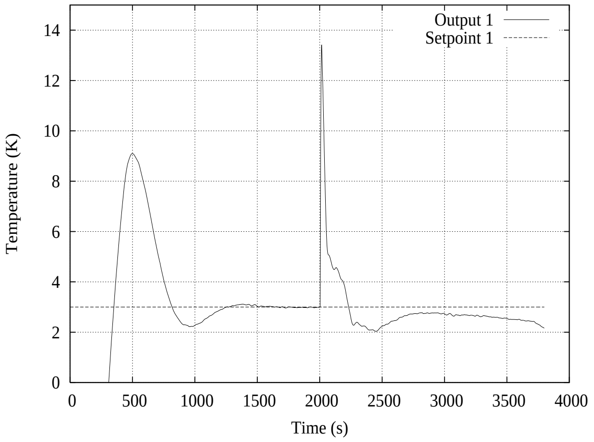

Figure 3. Estimated temperature of solids phase (K).

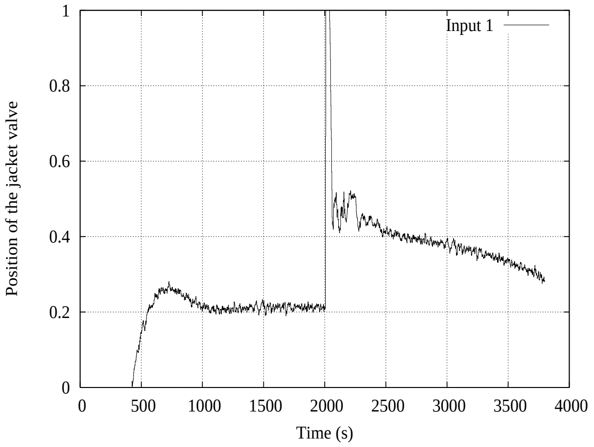

Figure 4. Position of the valve for the cold heat exchanger.

which acts as superviser of the complete process. The advanced control (nonlinear laws, state estimation) is performed by means of Fortran subroutines. Visual Basic interfaces and subroutines perform the communication between Eurotherm system and Fortran subroutines exploited as dll's.

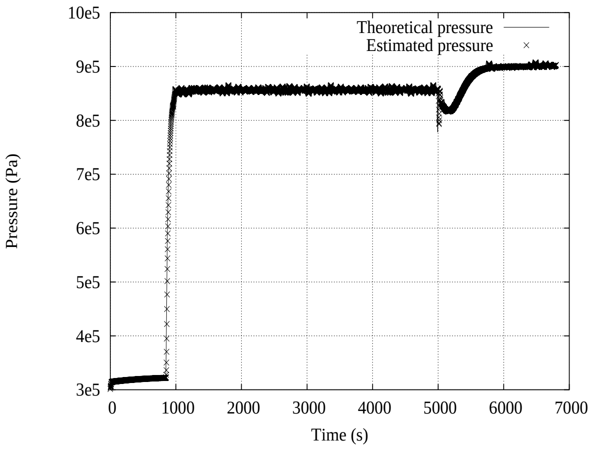

Figure 5. Estimated pressure of gas phase (Pa).

The reactor temperature (Figure 6) first reaches its set point and then the monomers are introduced. Around $t = 1300$s, the catalyst is introduced which provokes a rapid transient of the temperature which is after a short time again stabilized very closely to its set point. The valve of the cold heat exchanger (Figure 7) shows correspondingly its fast transient.

The first filtered pressure set point imposed around $t = 250$s is well followed by the reactor pressure (Figure 8). When reaction occurs, the pressure reaches rapidly its new set point in spite of a high demand on both monomers. Mass spectrometry allows us to verify with delay that the ratio of monomer concentrations was well maintained during the reaction.

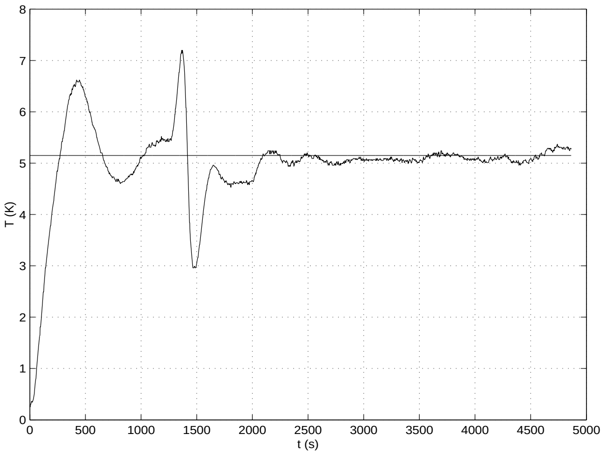

Figure 6. Measured temperature of the actual reactor (K) and set point.

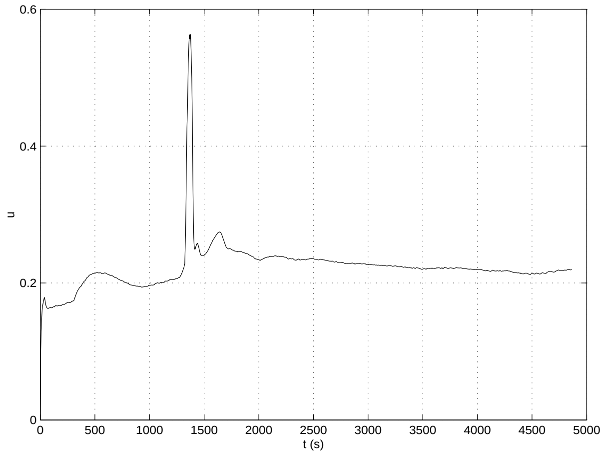

Figure 7. Position of the valve of the cold heat exchanger for the actual process.

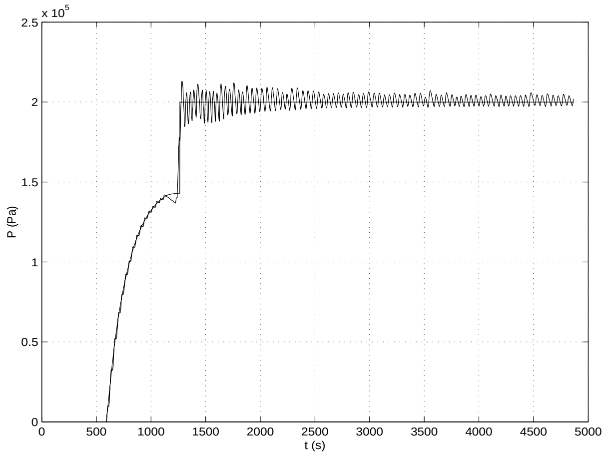

Figure 8. Measured pressure of the actual reactor (Pa) and set point.

4. CONCLUSION

Nonlinear control has been used in a multivariable context for a batch gas phase copolymerization reactor used for test of catalysts. It lies on a nonlinear model based on mass and energy balances including a growth model of polymer particle. The objective of the control to maintain the reactor temperature as close as possible to the set point is correctly respected on the actual reactor in spite of the highly exothermic reaction due to very reactive catalysts. The pressure also tracks very well its set point while the unmeasured ratio of monomer concentrations follows its requirements.

5. ACKNOWLEDMENTS

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