

Dynamic Behaviour and Control of an Industrial Fluidised-Bed Polymerisation Reactor

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Abstract

In gas-phase reactors for ethylene polymerisation, tight temperature control is of utmost importance to ensure that the temperature in the reaction zone is kept above the dew point of reactants, yet below the melt point of the polymer. Another important point is that if the temperature was under open-loop conditions, these reactors are prone to instability and limit cycles, however they can be easily stabilised with an appropriate temperature controller. In this work, a detailed study of the dynamic behaviour of an industrial fluidised-bed polymerisation reactor is carried out, with the location of bifurcation points and system stabilisation by controllers designed via optimisation in the frequency domain.

Keywords: stability, reactor temperature control, bifurcation

1. Introduction

There have been a great number of models proposed to describe the dynamic behaviour of the UNIPOL® process. Choi and Ray (1985) were the first to consider both emulsion and bubble phases in modelling the polyethylene fluidised-bed catalytic reactor. A review of dynamic behaviours and stability of polymerisation reactors has been provided by McAuley et al. (1995). They also demonstrate that the additions of the gas recycle system and heat exchange system to the reactor model make possible the study of complex dynamics and location of bifurcation points, which could not be reproduced just by the reactor kinetic model. In this work, the dynamic behaviour of the process, using a rigorous and validated model with industrial data, is analysed and an optimal design for the reactor temperature controller is proposed, using the cooling water valve opening as manipulated variable.

2. Process Model

A fluidised-bed zone and a disengagement zone compose the reactor, as shown in Figure 1. A heat exchanger is used to remove the reaction heat from the compressed recycle stream, and then the cooled gas is mixed with the feed stream to be re-injected in the base of the reactor. The solid catalyst (chromium based) is fed in a stream of nitrogen and then dragged to the fluidised-bed. The product is removed from the reactor

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by a discharge system operating in cycles determined by the production rate. In the disengagement zone the gas composition is analysed by chromatography.

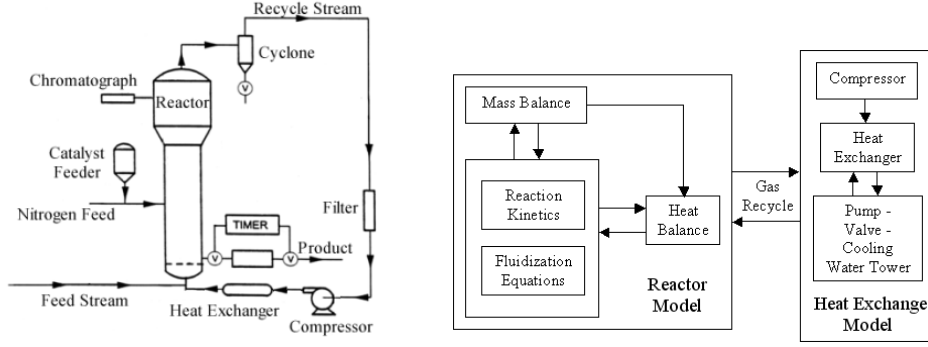


Figure 1. UNIPOL® process and system model

2.1 Reactor model

2.1.1 Reaction kinetics and fluidisation equations

The kinetics used in the model, given in Table 1, was developed by Gambetta et al. (2001) based on the works of McAuley (1991), McAuley et al. (1994), Xie et al. (1994), and Zacca (1995), for Ziegler-Natta catalyst system. Xie et al. (1994) pointed out that the same kinetic model could be used for chromium-oxide catalyst system.

In the reactor model it has been used the fluidisation equations of Kunni and Levenspiel (1991), from where Choi and Ray (1985) defined a basic set of equations, and then this set was used and adapted by Lageman (1989) and McAuley et al. (1994).

Table 1. Set of reactions used in the model.

Spontaneous site activation	$C_p \rightarrow P_0^k$	(1)
Chain initiation by monomer i	$P_0^k + M_i \rightarrow P_{\delta_i,i}^k$	(2)
Chain propagation by monomer j	$P_{n,i}^k + M_j \rightarrow P_{n+\delta_j,j}^k$	(3)
Spontaneous chain transfer	$P_{n,i}^k \rightarrow P_0^k + D_n^k$	(4)
Spontaneous chain deactivation	$P_{n,i}^k \rightarrow C_d + D_n^k$	(5)
	$P_0^k \rightarrow C_d$	(6)

where C_p is potential site, P_0^k is active site of type k , M_i is monomer of type i , $P_{\delta_i,i}^k$ is initiated chain of monomer type i and site type k , $P_{n,i}^k$ is live polymer with \bar{n} monomers, terminated in monomer type i and site type k , D_n^k is dead polymer with \bar{n} monomers of site k , and C_d is dead site. In this work, it has been used two types of monomers and two types of catalytic sites.

The distribution of polymeric chain lengths, responsible for keeping track of the mean properties of the polymer per type of site or end groups, including the mean molecular weights, polydispersion, number of active and dead polymer chains, are described by the above kinetic mechanism and the polymer moment definitions.

2.1.2 Mass balance and Heat balance

As stated by McAuley et al. (1994), the two-phase fluidised-bed model can be approximated by a well-mixed reactor, so the mass and heat balances use only one well-mixed zone, with a gas phase and a solid phase. The following gaseous components have been considered in the mass balance (Equation 7): ethylene, propylene, hydrogen, nitrogen, oxygen and inert saturated organic.

$$\frac{d[y]}{dt} = \frac{W_y - (Q_p \varepsilon_{mf} + Q_v)[y] - R_y}{V_g + V_d + V_b} \quad (7)$$

where y represents the gases components in the reactor. W_y , $[y]$, and R_y represents, respectively, the molar feed flow rate, the concentration and the rate of consumption of these gases, except for inert gases, where R_y is null. Q_p and Q_v are, respectively, the polymer product and the bleed stream flow rates. ε_{mf} is the bed porosity. V_g , V_d , and V_b are the volumes of gas phase, disengagement zone, and bubble phase, respectively.

The energy balance, neglecting the heat loss through the reactor wall, is given by Equation 8.

$$\frac{dT}{dt} = \frac{H_f + H_g - H_{out} + H_R}{m_s c_{ps}} \quad (8)$$

where $m_s c_{ps}$ is the thermal capacity of the system (reactor vessel and polymer in the bed), H_f and H_g are, respectively, the enthalpy of the fresh feed and the recycle stream entering the reactor. H_{out} is the enthalpy of the polymer product and the bleed stream leaving the reactor, and H_R is the reaction heat.

2.2 Heat exchanger model

An accurate representation of the heat removal system can be obtained using a staged heat exchanger model (Dadebo et al., 1997). The conventional log-mean temperature difference (LMTD) driving force model was used for the external single-pass shell and tube heat exchanger.

$$\frac{dT_{w,j}}{dt} = \frac{N F_w}{M_w} (T_{w,j-1} - T_{w,j}) - \frac{U A}{M_w c_{pw}} \Delta T_{lm,j}, \quad j = 1, \dots, N \quad (9)$$

$$\frac{dT_{g,j}}{dt} = \frac{N F_r}{M_g} (T_{g,j+1} - T_{g,j}) + \frac{U A}{M_g c_{pg}} \Delta T_{lm,j}, \quad j = 1, \dots, N \quad (10)$$

$$\text{where } \Delta T_{lm,j} = \frac{(T_{w,j} - T_{g,j+1}) - (T_{w,j-1} - T_{g,j})}{\ln \left(\frac{T_{w,j} - T_{g,j+1}}{T_{w,j-1} - T_{g,j}} \right)} \quad (11)$$

$\Delta T_{lm,j}$ is the log-mean temperature difference between the water and the gas in the j -th stage. M_w and M_g are the total molar holdups in the water and gas sides of the heat exchanger, respectively. F_w and F_g are the flow rate of water and gas, respectively. $T_{g,N+1}$ is the inlet gas temperature and $T_{w,0}$ is the inlet cooling water temperature to the external heat exchanger. $T_{g,j}$ and $T_{w,j}$ denote the temperature of the gas and coolant, respectively, leaving the j -th stage of the heat exchanger.

3. Dynamic Behaviour Analysis

The effects of the reactor operating conditions on the process dynamics and stability were analysed using the reactor model, including the recycle stream and the external cooler. The developed model was implemented in AUTO®, software for continuation and bifurcation problems in ordinary differential equations. Unstable steady states, limit cycles, and excursions toward unacceptably high-temperature steady states arise during the model simulation, when supposing the reactor operation with open-loop temperature control and the addition of a reactor total pressure controller to the system, whose manipulated variable is the ethylene feed flow rate. These non-linear dynamic behaviours, as shown in Figure 2, can be explained by positive feedback between the reactor temperature and the reaction rate.

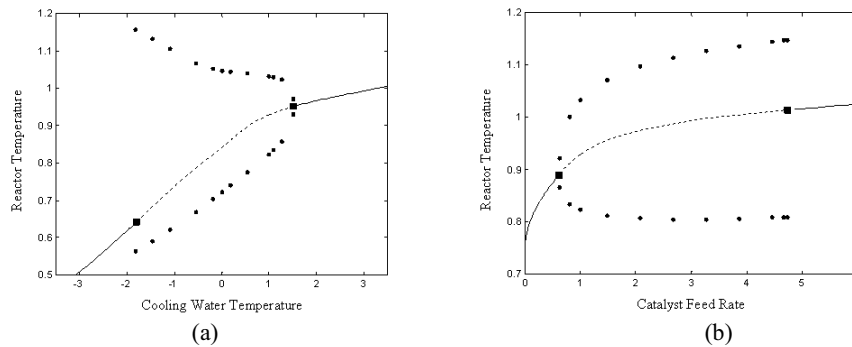


Figure 2. The effect of cooling water temperature (a) and catalyst feed flow rate (b) on stability: — stable steady state; --- unstable steady state; ■ Hopf bifurcation; ● stable limit cycle.

If the reactor temperature is above the unstable steady-state temperature, then the heat removal in the heat exchanger is larger than the steady-state heat-generation rate (McAuley et al., 1995). As result, the reactor temperature begins to decrease, decreasing the rate of reaction and the product outflow rate, thereby reducing the rate at which catalyst flows from the reactor. Thus, catalyst and monomer begin to accumulate in the reactor, increasing the temperature, the rate of reaction, and the product outflow rate, resuming the limit cycle. In Figure 3a, it can be shown the non-linear relationship between the reactor temperature and the cooling water valve opening. The gain in the reactor temperature is reduced as the valve moves in the fully open direction. Beside, after certain valve opening condition, heat exchange does not occur due to the cooling tower thermal limitation.

4. Process control

The reactor system is easily stabilised with a proportional controller. However, the addition of integral and derivative action eliminates offset and leads to faster controller performance (Dadebo et al., 1997). A PID temperature controller was designed using the SIOM-MMA (Sequential Iterative Optimisation Method - Multi-Model Approach, Faccin and Trierweiler, 2004), based on a frequency domain optimisation problem for a given desired performance. The method is able to design a PID controller for different operating points with robust performance, as shown in Figure 3b. Thus, different unstable regions of valve opening, shown in Figure 3a, were used in this method to the robust controller design. Satisfactory results were also obtained for rejecting disturbances.

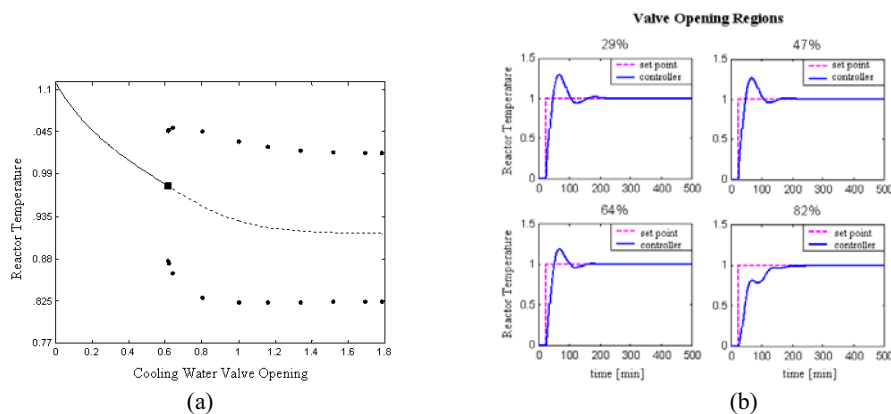


Figure 3. (a) The effect of cooling water valve opening on stability: — stable steady state; --- unstable steady state; ■ Hopf bifurcation; ● stable limit cycle. (b) Temperature response for a step setpoint change using the robust PID controller.

If the control valve saturates (a common situation in the industrial reactor), then the reactor operates without a feedback temperature controller, leading to oscillatory behaviour and limit cycles, as shown in Figure 4.

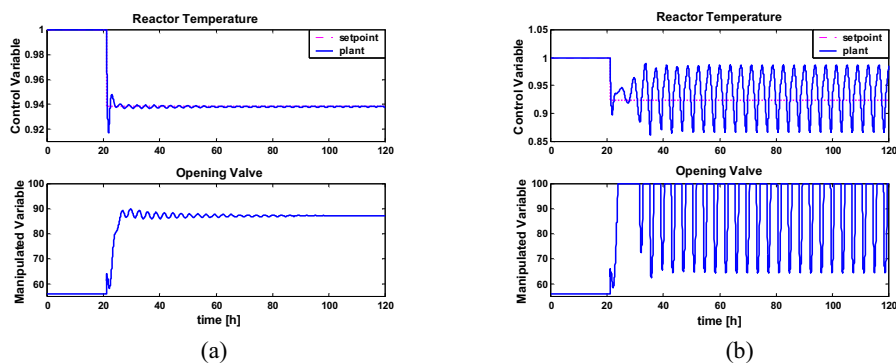


Figure 4. Temperature response for a step setpoint change: (a) leading to control valve fully open direction and (b) leading to control valve saturation.

Therefore, manipulation of valve opening alone is not sufficient to bring the temperature back to the desired level. The limit cycle can be removed from the system when auxiliary variables are used in the reactor temperature control. For instance, catalyst feed rate, inert saturated organic feed rate, and ethylene partial-pressure controller setpoint can reduce the rate of heat generation and stabilise the reactor temperature controller in the desired setpoint (Salau, 2004). However, all of these auxiliary variables reduce the production rate and other problems may arise with their use, some of them are shown in Table 2.

Table 2. Problems in using auxiliary variables to help control the reactor temperature.

Auxiliary Variables	Problems
Catalyst feed rate	It is difficult to determinate the flow in the feeder due to its small sensibility. High impact in the reactor production.
Inert saturated organic feed rate	If the mixture dew point were achieved, the distributed plate of gas in the rector will block. To prevent accumulation, it has to be removed from the reactor through the product or purge streams.
Ethylene partial-pressure controller setpoint	It causes a decrease in the recycle heat exchange capacity, by increasing the nitrogen/ethylene concentration ratio.

5. Conclusion

It was shown that control of gas-phase polymerisation reactors is a difficult task due to high non-linearity of the system and the strong interaction of the process variables. In the non-linear dynamic analysis, the complex behaviours were observed within the range of industrial operating conditions, with no modification in the kinetic parameters, as opposed to the existing works found in the literature. The use of an appropriate strategy for closing the reactor temperature control loop avoids the undesired non-linear dynamic behaviour.

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