UNISOTHERMAL CONTROL OF THE CASCADE OF CSTR USING CONSTANT DEGREE OF THERMAL STABILITY

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Unisothermal control of a cascade of continuous stirred tank reactors was analyzed. The control strategy is based on keeping the same degree of the thermal stability for all reactors in the cascade. The cooling driving force (the stationary temperature difference between the reaction mixture and the coolant) was used as the measure of the thermal regime stability. The structure of possible steady states was established, the thermal stability and technological properties of steady states were discussed. The most advantageous set of stationary states is recommended and the possibility of practical application of the suggested control strategy was discussed.

The previous paper[1] dealt with the control of degree of conversion in a cascade of continuous stirred tank reactors (CSTR) with an autocatalytic reaction under efficient independent temperature control. The reaction temperatures were supposed to be fixed and equal for all members of the cascade. Removal of reaction heat is the crucial problem in many industrial processes. For the temperature control to be reliable, the reactors operating states should have autoregulating properties, that means they should be stable from the point of view of their thermal regimes. In this paper a control strategy is suggested to ensure comparable degree of the thermal regime stability for all reactors in the cascade. The control strategy is applied to a cascade of polymerization reactors affected by a strong dependence of viscosity on degree of conversion and on temperature.

THEORETICAL

The mathematical model from the previous paper was used for simulations. The model of an ideally mixed vessel was used for both the reaction volume and the cooling system. The following equations were used for the simulations:
The key component balance

$$Fc_A(0)\,[X(I) - X(I-1)] + \nu_A r(I)\,V_r(I) = -V_r(I)c_A(0)\,\mathrm{d}X(I)/\mathrm{d}\tau \qquad (1)$$

Heat balance of the reaction mixture

$$F\varrho c_p[T(I-1) - T(I)] + r(I)\, V_r(I)\,(-\Delta H) + Q_{st} = \\ = K(I)\, A(I)\,[T(I) - T_c(I)] + V_r(I)\varrho c_p\, \mathrm{d}T(I)/\mathrm{d}\tau \qquad (2)$$

Heat balance of the cooling system

$$F_c\varrho_c c_{pc}[T_{ci}(I) - T_c(I)] + K(I)\, A(I)\,[T(I) - T_c(I)] = \\ = V_{cs}(I)\varrho_c c_{pc}\, \mathrm{d}T_c(I)/\mathrm{d}\tau \qquad (3)$$

The scheme of one reactor from the cascade is shown in Fig. 1.

Rate Equation

The rate equation of the second order reaction with the Arrhenius dependence of the rate constant was used[3,4].

$$r(I) = k \,.\, c_A(I)^2 \qquad (4)$$

$$k = k_0 \,.\, \exp\left[-E/(R \,.\, T(I))\right] \qquad (5)$$

Heat Transfer Coefficient and Heat Introduced by the Stirrer Motion

Exponential laws were applied to describe the effect of the degree of conversion and the effect of temperature on viscosity, heat transfer coefficient and heat introduced by the stirrer motion. The equations were given in the previous paper[1].

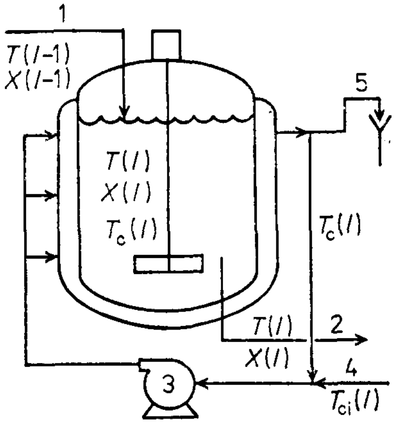

FIG. 1

Scheme of one member of the cascade. 1 Feed; 2 outlet; 3 recycling pump; 4 fresh coolant feed; 5 exit of the coolant

Determination of Steady States and their Stability

Graphical and numerical procedures were used to find out the steady states. The values of all heat balance items were expressed as functions of the reaction temperature. An example of the effect of the reaction temperature on heat balance is given in Fig. 2.

In order to evaluate the steady states stability, the trajectories of pseudostationary states in the phase plane X–T were constructed[2]. The values of the degree of conversion in the pseudostationary states were obtained by solving Eq. (*1*) introducing the condition of the stationary state.

$$\mathrm{d}X(I)/\mathrm{d}\tau = 0 \tag{6}$$

As regards the second order reaction, all pseudostationary states have stable orientation of the driving force of conversion changes. An example is given in Fig. 3. The temperature values in the pseudostationary states were obtained analogically by solving Eq. (*2*) for different degree of conversion using.

$$\mathrm{d}T(I)/\mathrm{d}\tau = 0 \tag{7}$$

The pseudostationary trajectory determines the orientation of temperature motion in the phase plane. The reaction temperature tends to move towards the stable pseudostationary states and out of the unstable ones.

The stationary states of the reactor were determined as the intersections of the temperature and conversion pseudostationary curves. Stability of the stationary states can be estimated from the orientation of the motion in its surroundings. The motion direction in the phase plane is determined by the vector, the components of which are the driving forces of the conversion and the temperature changes.

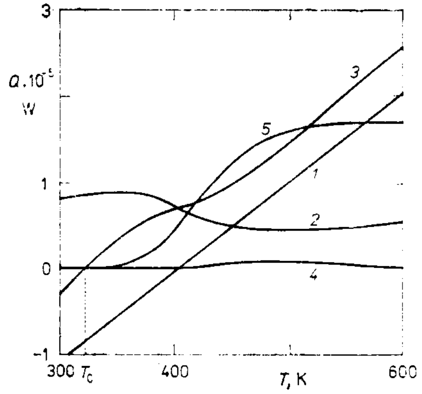

FIG. 2

An example of the structure of the reactor heat balance. *1* Rate of the feed cooling; *2* rate of the wall cooling under constant driving force; *3* overall rate of cooling; *4* rate of heat production by the stirrer motion; *5* overall rate of heat production; T_0 temperature of zero cooling rate

RESULTS AND DISCUSSION

Control Strategy

Removing of heat produced in the reactor is the key problem of many industrial processes regimes control. As to reactions in nonviscous media, the reaction heat is the prevailing item in heat production. As regards reactions in viscous media, e.g. polymerizations, heat introduced by the reactor stirrer motion may be important. The produced heat is supposed to be removed using two ways of cooling – by heat transfer through the reactor wall (wall cooling) and by cold feed of reaction mixture (feed cooling). The wall cooling is the prevailing way of heat removal in case of strongly exothermal reactions.

The total heat which must be removed can be estimated from the overall heat balance of the cascade

$$Fc_A(0)\,X(N)\,(-\Delta H) + \sum_{I=1}^{N} Q_{st}(I) = \sum_{I=1}^{N} \{K(I)\,A(I)\,[T(I) - T_c(I)]\} + F\varrho c_p\,[T(N) - T(0)]\,, \tag{8}$$

where N is sum of reactors in cascade, Q_{st} rate of heat production by the stirrer motion, $X(N)$ exit conversion at the end of the cascade, $T(N)$ temperature of the reaction mixture leaving the cascade, $T(0)$ temperature of the reaction mixture entering the cascade.

The heat balance in most chemical processes cannot be evaluated with sufficient accuracy so as the exit conversion could be controlled according to the heat balance. If the exit conversion is high (close to 1), the overall heat production is practically proportional to the feed rate F. An estimate of total heat Q_W, which must be transferred through the reactor walls, can be evaluated from the heat balance (*8*), where

$$Q_W = \sum_{I=1}^{N} \{K(I)\,A(I)\,[T(I) - T_c(I)]\}\,. \tag{9}$$

Two principles of temperature control can be applied to chemical reactors: (i) stabilization of inlet stream and parameters, (ii) a feedback control based on inlet variable manipulation. The stabilization of inlet streams is the simplest control mode in most of cases. Of course, the operation points must be open-loop stable for the stabilization to be applicable. In a cascade of reactors, the regimes of individual reactors may differ in stability due to different properties of the reaction mixture (degrees of conversion, viscosities, reaction rates). The control algorithm tested in this paper is based on the assumption, that all reactors should be operated with the same degree of the regime thermal stability.

There is no simple way for the quantitative characterization of the regime stability, so that some heuristic rules must be used to define the quantitative measure of stability.

Rule 1. The measure of the open-loop stability is the driving force of the wall cooling, that means the difference between the reaction temperature and the temperature of the cooling system. The aim of the regime optimization is to keep the driving force at the same set value in all reactors by manipulation of stationary reaction temperatures $T(I)$.

$$T(I) - T_c(I) = \text{const.} \qquad (10)$$

Rule 2. The measure of the regime stability is the temperature difference between the reaction temperature and the temperature of zero rate of cooling. The meaning of the temperature of zero cooling rate is illustrated by Fig. 2. At this temperature the sum of the wall cooling and the feed cooling is equal to zero. At this control mode the driving force of overall cooling is constant.

$$T(I) - T_0(I) = \text{const.} \qquad (11)$$

An estimate of the necessary cooling driving force can be obtained by solving Eq. (*9*). As the driving force is the decisive factor affecting the thermal regime stability, its evaluation indicates whether the control task is simple, difficult or impossible. The discussed control strategy must not be applied at least to one reactor for a degree of freedom to be obtained so as the heat balance could be kept in stationary state. For example, somewhat higher driving force than that calculated from Eq. (*9*) can be used in all reactors with the exception of the last one to remove most of reaction heat. The last reactor should be operated either at fixed coolant temperature for the rest of heat to be removed or under adiabatic regime that may be advantageous for reactions in viscous media.

Pathological Stationary States

Three types of stationary states can exist for the control strategy based on keeping the cooling driving force constant (Fig. 4). The stationary states differ in the prevailing mechanism of cooling and heat production.

Wall Cooling Prevails

Most of heat is produced by chemical reaction, the cold introduced by the feed is negligible, most of heat is removed through the wall. This type of steady state is the desired one from the technological point of view.

Feed Cooling Prevails

If heat transfer through the reactor wall is slow, the reaction heat cannot be removed by the wall cooling and most of heat must be used to heat the cold feed. The reactor operates in a pseudoadiabatic regime. This type of steady state cannot be accepted for the most processes, because the reaction temperature is too high.

Chemical Reaction Extinction

If the heat production rate is slow, higher amount of heat can be removed under the set driving force than can be produced by the reaction. Due to this fact the reaction temperature drops, the heat produced by reaction is negligible and, on the contrary, heat is introduced by the feed, temperature of which is higher than the stationary temperature in the reactor. Heat introduced by the feed is removed through the reactor wall.

All types of stationary states can be both stable and unstable, according to the set value of the cooling driving force. One or three states may exist in the case of second order reaction. When using the tested control strategy, all steady states were found to be stable due to the fact that the driving force of the wall cooling is limited.

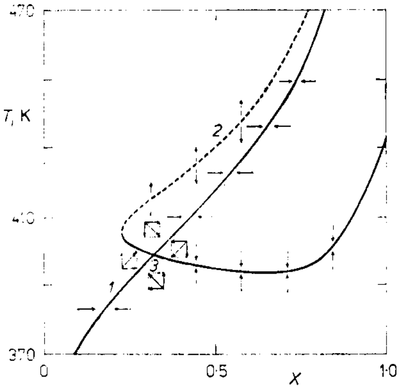

FIG. 3

An example of pseudostationary trajectories construction. *1* Trajectory of conversion pseudostationary states; *2* trajectory of temperature pseudostationary states (solid line – stable, dashed line – unstable states); *3* stationary state

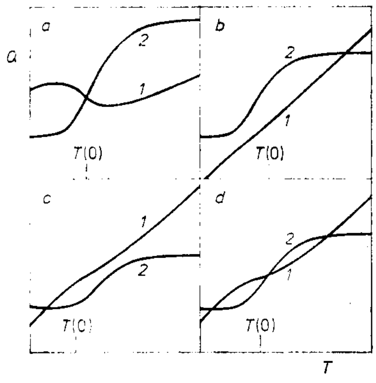

FIG. 4

Heat balance for different types of stationary states. *a* Desired steady state with intensive wall cooling; *b* pseudoadiabatic state, feed cooling prevails; *c* extinction of regime; *d* steady state multiplicity. *1* Overall rate of cooling; *2* overall rate of heat production; $T(0)$ temperature of the feed.

Reactions in Nonviscous Media

When applying the algorithms to a positive order reaction no problems are expected. Due to decreasing reactivity, the reaction temperature increases with the increasing number of reactor (Fig. 5). Problems with instabilities may appear in the first reactor if the rule 1 is used as the stability measure due to the fact that a very cold feed may enter the first reactor.

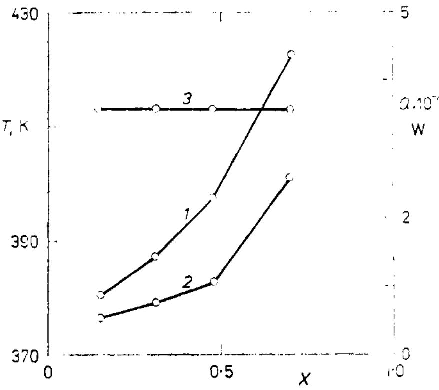

FIG. 5

An example of stationary states set for four member cascade with reaction in nonviscous medium. Rule 1; $T(I)$-$T_c(I) = 4{\cdot}8$ K; $T(0) = 375$ K; *1* reaction mixture temperature; *2* rate of the feed cooling; *3* rate of the wall cooling

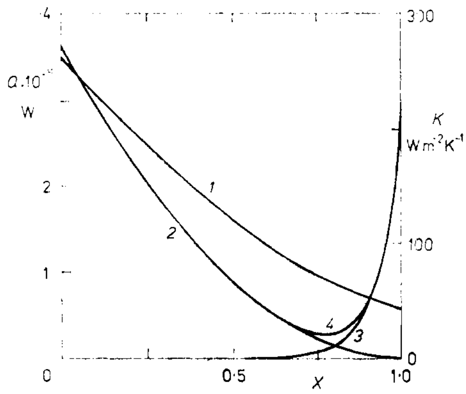

FIG. 6

Effect of degree of conversion on behaviour of a polymerization reaction mixture. $T = 410$ K; *1* heat transfer coeffficient; *2* reaction heat; *3* heat introduced by the stirrer motion; *4* overall heat production

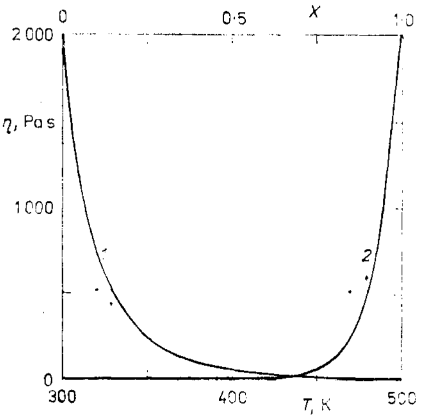

FIG. 7

An example of viscosity dependence on temperature and on conversion. *1* Effect of temperature ($X = 0{\cdot}7$); *2* effect of conversion ($T = 413$ K)

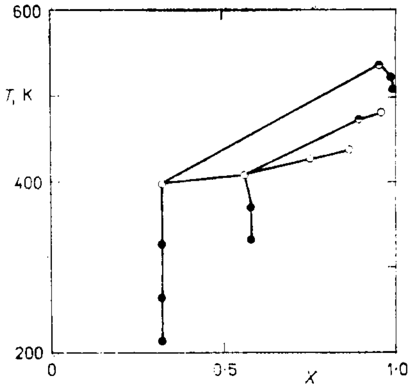

FIG. 8

An example of stationary states set for four member cascade with constant cooling driving force. $T(I)$-$T_c(I) = 12$ K; $T(0) = 398{\cdot}74$ K; ○ desired steady state; ◒ pseudodiabatic state; ● extinction of regime

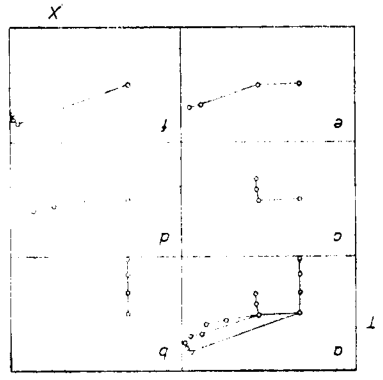

FIG. 9

All branches of stationary states for four member cascade with constant cooling driving forces; a all stationary states; b–f individual branches. $T(I)$-$T_c(I) = 12$ K; $T(0) = 398{\cdot}74$ K

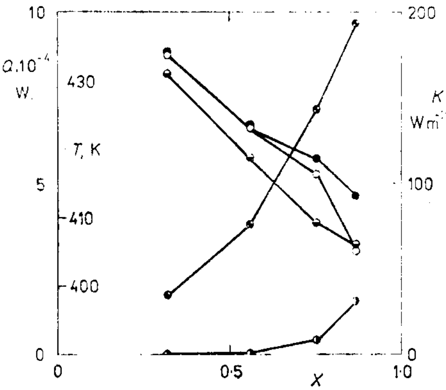

FIG. 10

An example of optimal stationary states set for four member cascade. $T(I)$-$T_c(I) = 12$ K; $T(0) = 398{\cdot}84$ K; ⊗ reaction mixture temperature; ⊖ heat transfer coefficient; ○ reaction heat; ◑ heat introduced by the stirrer; ● overall heat production

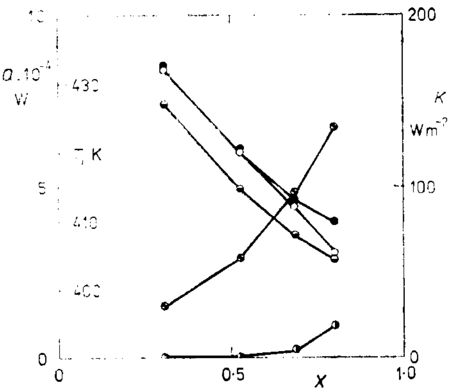

FIG. 11

An example of stationary states set for four member cascade. Rule 2; $T(I)$-$T_0(I) = 10$ K; $T(0) = 395$ K; ⊗ reaction mixture temperature; ⊖ heat transfer coefficient; ○ reaction heat; ◑ heat introduced by the stirrer; ● overall heat production

Polymerization Reactors

Behaviour of polymerization reactors is complicated by the effect of conversion and temperature on the reaction mixture viscosity. Examples of the effect are given in Figs 6 and 7. Rate of the heat transfer through the wall decrease and amount of heat introduced by stirrer motion increases with increasing viscosity. The control algorithm tends to fail in the reactors with high degree of conversion.

An example of the whole structure of stationary states for four member cascade is given in Fig. 8. It can be seen that the multiple stationary states exist when using the control rule 1. As each multiplicity causes multiplicities of states in all reactors downstream, a complicated structure of the regime branches is formed. All possible stationary branches for chosen cooling driving force are shown in Fig. 9. The branch *d* is the most advantageous from the technological point of view because the wall cooling prevails in all the employed stationary states (Fig. 4*a*). Values of some important quantities for this case are given in Fig. 10.

The cooling driving force is not an exact quantitative measure of the open loop stability. But the advantage of the control strategy using constant cooling driving force consists in that the practical application is based on directly measured quantities such as the reaction mixture and cooling system temperatures.

A more sophisticated control strategies, e.g. that one using the temperature of zero cooling rate (rule 2), are more difficult to be applied. An example of stationary states set fulfilling the control rule 2 is given in Fig. 11. To evaluate the temperature of zero cooling rate, the reaction mixture feed rate and inlet temperature, the heat transfer coefficient, and the coolant temperature have to be measured on line.

CONCLUSION

The analysis has proved that keeping the driving forces of the wall cooling at a constant value in all cascade reactores may represent a useful strategy for temperature control. By selecting a proper value of the driving force, thermally stable steady states can be achieved in all cascade members. This control strategy is efficient at lower degrees of conversion (at low viscosity of the reaction mixture). At high degrees of conversion (high viscosity of the reaction mixture) raises the heat transfer resistance and the heat flux through the reactor wall is small. As a consequence, pathological steady states may exist along with the desired ones. A transfer into these pathological states must be avoided.

SYMBOLS

A	heat transfer area, m^2
c_A	concentration of reactant, mol m^{-3}
c_p	specific heat capacity of reaction mixture, J kg^{-1} K^{-1}

c_{pc}	specific heat capacity of coolant, J kg^{-1} K^{-1}
E	activation energy, J mol^{-1}
F	volume flow rate of reaction mixture, m^3 s^{-1}
F_c	volume flow rate of coolant, m^3 s^{-1}
ΔH	reaction enthalpy, J mol^{-1}
I	ordinal number of reactor in the cascade
K	heat transfer coefficient, W m^{-2} K^{-1}
k	rate constant, m^3 mol^{-1} s^{-1}
k_0	constant in Eq. (*5*), m^3 mol^{-1} s^{-1}
N	total number of reactors in the cascade
Q_W	heat flux through the reactor wall, W
Q_{st}	amount of heat introduced by the stirrer motion, W
r	reaction rate, mol m^{-3} s^{-1}
R	universal gas constant, J mol^{-1} K^{-1}
T	temperature of the reaction mixture, K
T_c	exit temperature of coolant, K
T_{ci}	inlet temperature of coolant, K
T_0	temperature of zero cooling rate, K
V_{cs}	volume of cooling system, m^3
V_r	volume of the reaction mixture, m^3
X	degree of conversion
η	viscosity of reaction mixture, Pa s
ν_A	stoichiometric coefficient of reactant
ϱ	density of the reaction mixture, kg m^{-3}
ϱ_c	density of coolant, kg m^{-3}
τ	time, s

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