TEMPERATURE CONTROL IN CHEMICAL REACTORS WITH SLOW RESPONSE OF THE COOLING SYSTEM; DEFINITION OF COEFFICIENTS OF CONTROL SAFETY

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Coefficients of static, dynamic and information safety are defined. They could be used for consideration of how difficult is the feedback of temperature control in a chemical reactor with exothermic reaction. An analysis is made for a batch reactor which should be kept in a pseudo-stationary state which is unstable at the open control loop. Control is based on measurement of the reaction mixture temperature and on evaluation of temperature derivative with time. The action quantity is flow rate of the carrier in the cooling system. The result of an analysis is the highest operating temperature of the mixture at which the safety of the reactor operation is still secured and further the trajectory on which it is possible to reach in the shortest time the given operating temperature at preserving safety of the reactor operation.

In temperature control of the reaction mixture in reactors with exothermic reaction the most important goal is control of the safe operation, i.e. elimination of the uncontrolled temperature rise. It is simultaneously required to make use of the highest possible reaction temperature as the reactor output is usually greater at higher temperatures. As the danger of uncontrolled temperature rise increases with increasing reaction temperature it is necessary to make a compromise between the requirements on safe operation and the high output. Problems with the temperature control are of importance especially with high-capacity reactors which have a disadvantageous ratio of their surface to volume. It is sometimes necessary in these reactors to make use of operating states which are at the open control loop unstable[1-2]. Necessary condition of temperature control in these states is the use of the feedback controller. Literature on control of chemical reactors is relatively rich[4-5]. But majority of studies is devoted to problems of multiple states and qualitative analysis of stable states on systems with stabilised inlets (in autonomous systems). Qualitative analysis of stability has a decisive significance in the case when temperature in the reactor is controlled by stabilisation of inlet streams. If the temperature is to be controlled by use of the feedback circuit the qualitative result of stability analysis (stable-unstable) loses its basical significance. The analysis of how difficult is the temperature control must be based on a qualitative analysis of dynamic properties of the system.

In this study is proposed a method of analysis of safety of feedback temperature control making use of the mathematical reactor model and evaluation of motion trajectory in the phase plane.

THEORETICAL

Definition of Safety Coefficients

At evaluation of safe temperature control it is necessary to take care of three aspects: static safety, dynamic safety and safe information on the state of system.

Coefficient of static safety is defined as ratio of the highest cooling rate which can be reached and the rate of heat origin by chemical reaction at the given temperature of the reaction mixture. It is assumed that the cooling system is in steady state when

$$S_{st} = r_{c,max}/r_h .$$ (1)

Coefficient speaks out whether the reactor at the given temperature has the necessary cooling capacity so that it would be possible to remove the heat of reaction by the same rate as it originates.

Coefficient of dynamic safety is defined as the ratio of critical temperature rise and the momentous observed temperature rise at the given temperature of the mixture

$$S_{dyn} = (dT/dt)_{max}/(dT/dt)_{obs} .$$ (2)

The critical temperature rise is defined on basis of the following model. Let us assume that at the given temperature of the mixture is the cooling system set into operation at the highest possible rate. This means that the lowest temperature and the highest flow rate of the heat carrier are used. Temperature dependence of the mixture is the result of control action and depends on the state of cooling system in the moment of its performance. Beginning of the response is characterised by value of the observed temperature rise $(dT/dt)_{obs}$. The critical temperature rise $(dT/dt)_{max}$ is the highest temperature rise for which the response is still not classified as its failure from the technological point of view. Its failure can have two causes. At first a certain highest permitted temperature is given surpassing of which causes losses (*e.g.* quality of the product deteriorates) or there incurrs an uncontrolled rise in temperature of the mixture. Discussion considers only the second cause.

Coefficient of information safety is characterising the effect of noise in measurements of temperature of the mixture. The noise in temperature measurements aggravates first of all determination of the value of derivative of mixture temperature with time. Uncertainty in determination of the derivative is described by the width of the noise band of derivative $(dT/dt)_n$. Coefficient of information safety is then defined as the ratio of criticaly temperature rise and band width of noise derivative at the given mixture temperature

$$S_{inf} = (dT/dt)_{max}/(dT/dt)_n .$$ (3)

Reactor Model

Let us assume that it is possible to describe the state of the reactor by three state quantities, *i.e.* by temperature of the reaction mixture, mean temperature of the heat carrier in the cooling system and degree of conversion of the key reaction component. Dynamic behaviour of the batch reactor is then described by the system of equations

$$dx/dt = r/c_{A0} \tag{4}$$

$$dT/dt = T_{ad}(r/c_{A0}) - A(T - T_c) \tag{5}$$

$$dT_c/dt = AB(T - T_c) - C(T_c - T_{ci}) \tag{6}$$

with the initial condition in the form

$$t = 0 \quad x = 0, \ T = T_0, \quad T_c = T_{c0}.$$

Numerical calculations of examples were performed at the assumption that the reactor cooling system is described by the model of a perfectly mixed vessel. For description of dependence of the reaction rate on degree of conversion and on temperature the data have been used for a reaction studied in the last study[3] in which it has been determined that the reaction has an autocatalytic character. In the range of conversion degrees from 0 to 0·15 it is possible to use the relation

$$T_{ad}(r/c_{A0}) = 9 \cdot 10^{11} (1 - 1·6296x) \exp(-10\,817.3/T), \tag{7}$$

for higher degrees of conversion the relation

$$T_{ad}(r/c_{A0}) = 4·6923 \cdot 10^{12} (1 - x^{1·87}) x^{0·85} \exp(-1\,0817·3/T). \tag{8}$$

Adiabatic increase in temperature due to reaction T_{ad} is 130 K, inlet temperature of the heat carrier into the cooling system T_{ci} is 283 K, parameter $A = 1·5 \cdot 10^{-4} \cdot s^{-1}$, parameter $B = 3·86 \, s^{-1}$ and parameter $C = 1·3 \cdot 10^{-3} \, s^{-1}$.

RESULTS AND DISCUSSION

Safe temperature control of the reaction mixture by use of the mathematical model of the system depends on reliability of information on the system. For control of industrial reactors the mixture temperature is usually measured continuously without significant delay. Degree of conversion is determined periodically, frequently with delay due to time necessary for performing the analysis. Sometimes the degree of conversion is not determined at all and it is then necessary to estimate it by calculation

e.g. from the heat balance. It is useful to divide uncertainties in determination of the system state into four groups:

I) Noise in temperature measurements of the mixture and in determination of derivatives of temperature.

II) Uncertainty in determination of the degree of conversion and related uncertainty in determination of reactivity of mixture, uncertainty in determination of inlet temperature and flow rate of heat carrier.

III) Uncertainty introduced by other not measured parameters, *e.g.* by change in the catalyst activity, scaling of heat transfer areas, random changes in inlet composition of the mixture and the change in flow rate of the heat carrier.

IV) Inaccuracy in structure of mathematical model of the system, *i.e.* effect of simplifications introduced into the model (assumption of ideal mixing, plug flow *etc.*).

Control analysis is limited to two-position (bang-bang) temperature control by flow rate of the heat carrier. Discussion is divided into two parts: *1*) Keeping the thermal balance of the reaction mixture in pseudostationary state, i.e. in the state when heat of reaction is removed at the same rate as it originates. Condition of pseudostationary state results from Eq. (5) by substitution of equality $dT/dt = 0$. *2*) Starting of the process, *i.e.* transition from initial state of the system into pseudostationary state.

Determination of the highest pseudostationary temperature of reaction mixture in the phase plane $T - T_c$. Fundamental for the pseudostationary state at two-position control is the alternate motion of temperature up and down in vicinity of the pseudostationary state. Since the switch-over cycles are short as compared to the time of duration of the whole batch it can be assumed at description of motion in vicinity of the pseudostationary state that reactivity of the mixture does not change. This means that is it possible to neglect the effect of conversion degree and changes of not measured parameters. The reaction rate then depends only on temperature.

With reactors where a strongly exothermic reaction takes place the limiting factor is the highest attainable cooling rate. Analysis for determination of the highest safe pseudostationary temperature is thus based on the analysis of behaviour for the highest cooling rate. Static limitation of the highest pseudostationary temperature is determined by intersection of the curve of pseudostationary states of the mixture and the curve (straight line) of pseudostationary states of the cooling system (Figs 1 and 2). The dynamically safe region of phase plane, at elimination of noise in temperature measurement of the mixture, is limited by two curves. First of all it is the curve (straight line) of pseudostationary states of the cooling system. This curve is defined by Eq. (6) by substituting of the condition

$$dT_c/dt = 0 , \quad T_c = T_{cs} .$$

Region of the phase plane, for which there holds $T_c < T_{cs}$ then cannot be reached. To reach the phase plane it would be necessary to subcool the heat carrier in the cooling system by another cooler medium. Moreover it is limited by the critical trajectory (separatrix). From the dynamic point of view the temperature control is safe when it is possible to ensure that after switch-over to the highest cooling rate, the temperature response of the mixture would not exceed the region of temperature decrease $(dT/dt < 0)$ and in the region of temperature increase $(dT/dt < 0)$ when the trajectory of response is passing through the pseudostationary state (singular point). The critical trajectory can be determined by numerical integration of the system of Eqs (5) and (6) with negative sign for time and with the use of the initial condition in the form

$$t = 0 \quad T_{c0} \to T_{cs}^+, \quad T_0 \to T_s^-.$$
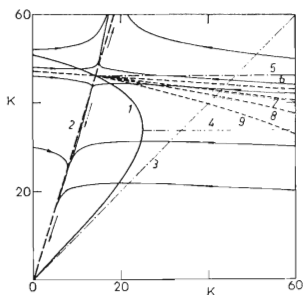

FIG. 1

Trajectory of temperature response of reaction mixture after switch-on of cooling in phase plane temperature of mixture in reactor — temperature of heat carrier in cooling system. $x = 0$; 1 Pseudostationary state of mixture, 2 pseudostationary state of cooling system, 3 curve of dependence $T = T_c$, 4 curve limiting region of stable and unstable states at open control loop, 5 $S_{s1} = 1$, 6 separatrix ($S_{dyn} = 1$), 7 $S_{dyn} = 2$, 8 $S_{dyn} = 3$, 9 $S_{dyn} = 5$
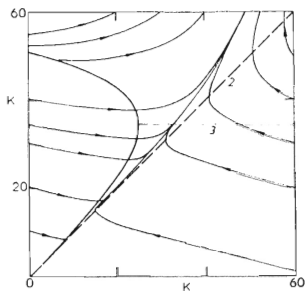
FIG. 2

Trajectory of temperature response of reaction mixture after switch-off of the cooling for the case given in Fig. 1. 1 Pseudostationary state of reaction mixture, 2 pseudostationary state of cooling system, simultaneously the dependence $T = T_c$, 3 curve limiting the region of stable and unstable reactor states

The + sign means that the value is being approached from the right (it is larger), − sign means that the value is being approached from the left (it is smaller). On the critical trajectory (Fig. 1) the coefficient of dynamic safety has the value $S_{dyn} = 1$. Temperature derivative of the mixture by time corresponds to the highest permitted temperature rise. In Fig. 3 is given the dependence of coefficients of static and dynamic safety and of critical temperature rise on temperature of the mixture (it is assumed that the effect of degree of conversion is negligible).

In the system with the noise effect the highest safe pseudostationary temperature of the mixture is limited by noise. For the safe control the noise in determination of temperature derivative must be smaller than the critical temperature rise. The limiting value corresponds to the case when $S_{inf} = 1$. This means the case when it is just possible to recognize in the noise that the temperature rise has reached the critical value. Of course in practice it is not admissible to undertake the risk of a failure resulting from stochastic effects in the system and so it is necessary to select much higher value (Fig. 3).

In majority of reactors there take place changes of not measured parameters. In Fig. 4 is given an example of calculation of pseudostationary temperatures of mixture and cooling rystem and critical trajectory for changes in mixture reactivity within the range $\pm 10\%$. The result demonstrates that it is necessary for temperature control in unstable state to count with a large parametric sensitivity of safety coefficients. As
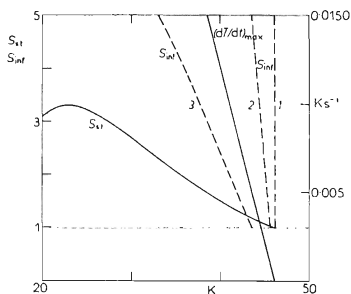

FIG. 3

Safety coefficient and critical temperature rise of reaction mixture in dependence on temperature of reaction mixture for the case given in Fig. 1. *1* $(dT/dt)_n = 0$, *2* 0·001, *3* 0·005
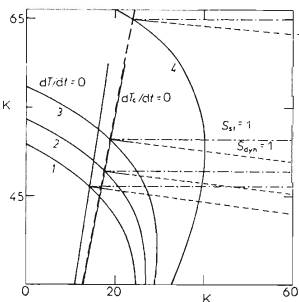
FIG. 4

Effect of conversion degree on pseudostationary reactor state and on critical trajectory. *1* $x = 0$, *2* $x = 0·495$ (change in mixture reactivity for 10%), *3* $x = 0·6$ (change in mixture reactivity for 20%), *4* $x = 0·8$

fluctuations of reactivity within the range of 10% relative are common, application of the forward nonadaptive control is limited (data included into the model of the system represent the forward control).

With reaction of the nonzero order position of pseudostationary temperature as well of critical trajectory changes with the degree of conversion. When it is prescribed to perform the reaction isothermally, it is necessary to choose the temperature so that safety at the highest mixture reactivity is secured (in majority of reactions this is at the beginning of the process). When it is admissible to perform the reaction nonisothermally, it is possible to vary the mixture temperature so that safety during the batch operation is constant. Examples of temperature dependence of the mixture, at constant safety coefficient, on degree of conversion for the studied reaction are given in Figs 4 to 6.

For fast reacting cooling systems is the highest permissible pseudostationary mixture temperature limited by static safety. Our experience and previous results outline that in an actual situation it is necessary to choose the coefficient of static safety even at adaptive control at least equal to 1.2. At lower values the control can fail. In reactors with large thermal capacity of the cooling system as the limiting factor can be the coefficient of informative safety.
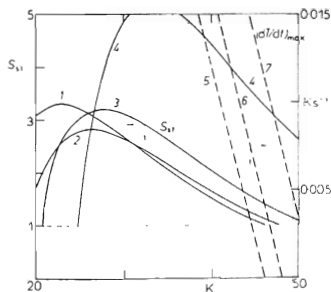

FIG. 5

Effect of conversion degree on dependence of coefficient of static safety and critical temperature rise on temperature of the reaction mixture. Solid line denotes dependence of S_{st}, dashed line $(dT/dt)_{max}$; 1 and 5 $x = 0$, 2 and 6 $x = 0.495$, 3 and 7 $x = 0.6$, 4 $x = 0.8$
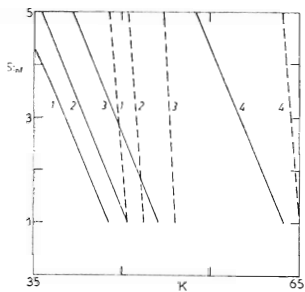
FIG. 6

Effect of conversion degree on dependence of coefficient of information safety on temperature of reaction mixture. Solid line denotes dependence for $(dT/dt)_n = 0.005$, dashed line for 0.001; other symbols used are the same as those in Fig. 5

Transformation of trajectories into the phase plane $T - (\mathrm{d}T/\mathrm{d}t)$. When the reactor is controlled only on basis of measurements of mixture temperatures it is advantageous to transform all important curves in the phase plane $T - T_c$ into the phase plane $T - (\mathrm{d}T/\mathrm{d}t)$. Interpretation of curves is given in Figs 7 and 8.

Starting of operation. Let us assume that it is possible to use at starting of temperature control two heat transfer media, the heating medium with inlet temperature T_{ch} and the cooling medium with inlet temperature T_{ci}. To the fastest start corresponds heating of the mixture to trajectory with the given value of coefficient of dynamic safety. After reaching this trajectory it is necessary to arrange for a motion along it by alternate opening and closing the flow of heat transfer medium into the cooling system (at two-position control). Examples of trajectories with constant dynamic (information) safety are demonstrated in Fig. 9.

The given value of coefficient of dynamic safety is fixed by noise and other sources of uncertainties in determination of the state of the system. The value of the coeffi-
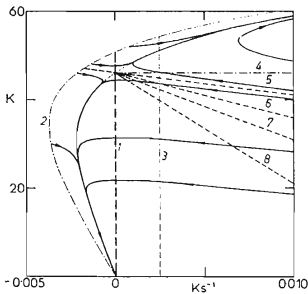

Fig. 7

Trajectory of temperature response of reaction mixture after switch-on of the cooling in the phase plane mixture temperature in reactor-derivative of mixture temperature by time for the case given in Fig. 1. *1* Pseudostationary states of reaction mixture, *2* pseudostationary states of the mixture for $T_c = T_{ci}$, *3* $(\mathrm{d}T/\mathrm{d}t)_n = 0.0025$, *4* $S_{st} = 1$, *5* separatrix $(S_{dyn} = 1)$, *6* $S_{dyn} = 2$, *7* $S_{dyn} = 3$, *8* $S_{dyn} = 5$
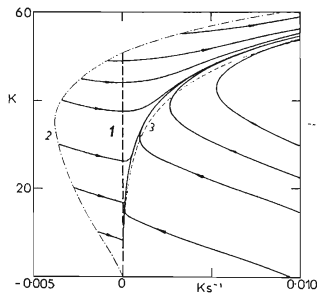
Fig. 8

Trajectory of temperature response of reaction mixture after switch-off of the cooling in the phase plane mixture temperature in the reactor-derivative of mixture temperature by time for the case given in Fig. 2. *1* Pseudostationary states of reaction mixture, *2* pseudostationary states of mixture for $T_c = T_{ci}$, *3* pseudostationary states of the reaction mixture for $T = T_c$

cient of dynamic safety must be selected so as to have the coefficient of dynamic safety always larger than one. Simultaneously it must be chosen so that the value of dynamic safety coefficient would not decrease below the value one due to uncertainty in determination of parameters of the system. The less accurate is the model the higher safety must be chosen.

Simulation of temperature control of the mixture. Finally, examples of control simulation are given, whose aim is to keep the coefficient of information safety at constant value.

After heating of the mixture to the temperature at which is the safety coefficient just equal to the given value, for a zero conversion degree of the key reactant, cooling of the reactor is switched on and a sample withdrawal for an analysis is simulated for determination of the momentous value of conversion degree. Cooling of the reactor is switched on for time t_a, corresponding to the expected time of analysis. Simultaneously is being evaluated the derivative of mixture temperature by time on the basis of "two-points" difference scheme. For momentous conversion degree, is cal-
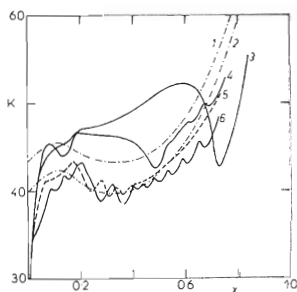

Fig. 9

Control simulation based on keeping constant the value of information safety. The effect of time for analysis of the mixture on control dependence. $S_{st} = 1·2$, $(dT/dt)_n = 0·0025$, $T_{ch} = 353$ K; *1* trajectory for $S_{inf} = 2$, *2* $S_{inf} = 5$, *3* trajectory of simulated control for $S_{inf} = 2$, $t_a = 200$, *4* $S_{inf} = 2$, $t_a = 600$, *5* $S_{inf} = 5$, $t_a = 200$, *6* $S_{inf} = 5$, $t_a = 600$
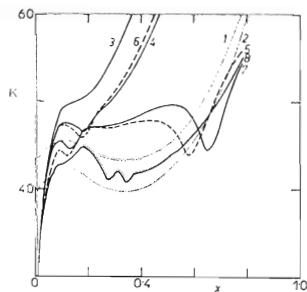
Fig. 10

Simulation of control based on keeping the information safety at constant value. Effect of changes in mixture reactivity on control dependence. $S_{st} = 1·2$, $T_{ch} = 353$, $(dT/dt)_n = 0·0025$, $t_a = 200$; *1* trajectory for $S_{inf} = 5$, *2* $S_{inf} = 5$, *3* trajectory of simulated control for $S_{inf} = 2$, uncertainty in evaluation of mixture reactivity in the model 20%, *4* $S_{inf} = 3$, 20%, *5* $S_{inf} = 5$, 20%, *6* $S_{inf} = 2$, 10%, *7* $S_{inf} = 3$, 10%, *8* $S_{inf} = 5$, 10%

culated the new mixture temperature by use of the mathematical model of the system which corresponds to the fixed safety coefficient. At cooling of the reactor being switched off or on then proceeds the temperature control up to the moment of reaching the calculated value of the mixture temperature. The whole procedure is then repeated.

At simulated control of the mixture temperature have been studied both the effect of time needed for determination of conversion degree by analysis (Fig. 9) and the effect of changes in mixture reactivity (accuracy of the mathematical model of the system, Fig. 10) on control behaviour. From simulation calculations results that in the case when the cooling system of the reactor has a large inertia and there is a noise in temperature measurements of the mixture it is necessary to fix the value of the coefficient of dynamic safety from three to five for securing of the control safety.

LIST OF SYMBOLS

$A = k_h P/(V c_p \varrho)$ parameter characterising heat transfer intensity between the reaction mixture in the reactor and heat transfer carrier in the cooling system (s^{-1})

$B = V c_p \varrho/(V_c c_{pc} \varrho_c)$ ratio of thermal capacities of reaction mixtures in the reactor and heat carrier in the cooling system

$C = F_c/V_c$ reciprocal residence time time of heat carrier in the cooling system (s^{-1})

c_{A0} initial concentration of key component $(kmol\ m^{-3})$

c_p, c_{pc} specific heat of reaction mixture and heat carrier $(kJ\ kg^{-1}\ K^{-1})$

F_c volumetric flow rate of hear carrier into the cooling system $(m^3\ s^{-1})$

k_h heat transfer coefficient $(kW\ m^{-2}\ K^{-1})$

P heat transfer area (m^2)

R gas law constant

r reaction rate $(kmol\ m^{-3}\ s^{-1})$

r_c, r_h cooling rate and rate of heat liberated by reaction $(K\ s^{-1})$

T temperature of reaction mixture (K)

T_{ad} adiabatic temperature rise (K)

T_{ch} temperature of heat carrier at the inlet into the cooling system at heating of the reaction mixture ιK)

T_c temperature of heat carrier in the cooling system (K)

T_{ci} temperature of heat carrier at the inlet into the cooling system at cooling of the reaction mixture (K)

T_0, T_{c0} initial values of quantities (K)

T_s, T_{cs} values corresponding to pseudostationary states (K)

t reaction time (s)

t_a time of analysis for determination of conversion degree (s)

S safety coefficient (Eqs (1) to (3))

V volume of reaction mixture in reactor (m^3)

V_c volume of heat carrier in cooling system (m^3)

x conversion degree of key component

ϱ, ϱ_c density of reaction mixture and heat carrier $(kg\ m^{-3})$

Subscripts

max largest value of quantity

obs observed value of quantity

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