
**NONISOTHERMAL CONTROL OF BATCH REACTOR
WITH REACTION BY DIGITAL COMPUTER.
EXPERIMENTAL VERIFICATION OF THE CONTROL ALGORITHM
BASED ON MATHEMATICAL MODEL OF THE SYSTEM**

František JIRÁČEK and Josef HORÁK

*Department of Organic Technology,
Institute of Chemical Technology, 16628 Prague 6*

Received on June 8th, 1982

Two-position feedback temperature control of the mixture in a batch reactor with the strongly exothermic autocatalytic reaction is studied. The control is based on continuous measurement of reaction temperature, evaluation of its time derivative and periodical determination of conversion of the reactant. On basis of numerical solution of equations of the mathematical model of the reactor is then evaluated the highest operating temperature of the mixture at which is still secured a safe operation of the reactor and trajectories along which the given operating temperature could be reached in the shortest time period. The manipulated variable was the flow rate of heat carrier into the cooler. Sampling of the reaction temperature and two-position switching over of flow rate of the heat carrier was performed in real time by use of the digital measuring center Hewlett-Packard 3052A. In the experiments has been verified the effect of changes in reactivity of the mixture, accuracy of the mathematical reactor model and effect of additional noise in the measured reaction temperature on control safety. Results of the experiments have proved that the proposed control algorithm enables safe control of the reaction temperature also in cases when the operating reactor states are unstable at the open control loop and when the cooler has a slow response to changes in the manipulated variable.

In the previous study¹ a method has been proposed of analysis of safe feedback temperature control of the reaction mixture making use of the mathematical model of the reactor and evaluation of motion trajectory in the phase plane. The method enables to determine by use of the coefficients of static, dynamic and information safety the highest pseudostationary temperature of the mixture at which the operating safety of the reactor is still preserved and simultaneously evaluate trajectories on which it is possible to reach the required temperature in the shortest time interval.

The aim of this study is experimental verification of the method for temperature control in a batch reactor with strongly exothermic reaction. The reactor has to be kept in unstable pseudostationary state at open control loop under conditions that the cooler of the reactor has a slow response to changes in the manipulated variable.

THEORETICAL

The basis of the proposed method is described in detail in the previous paper¹, where the temperature control is performed along the trajectory of constant coefficients of static, dynamic and information safety. So is simultaneously determined that the temperature is kept at the highest value at preserved safe reactor operation. Determination of the highest pseudostationary reaction temperature on basis of given safety coefficients is based on numerical solution of equations of the mathematical reactor model. Under the assumption that in vicinity of the pseudostationary state can be neglected the effect of changes in reactivity of the mixture *i.e.* the effect of changes in degree of conversion of the key component and changes in measured parameters, dynamic behaviour of the batch reactor is described by the system of equations

$$dT/dt = T_{ad}r/c_{A0} - A(T - T_c) - A_z(T - T_z) \quad (1)$$

$$dT_c/dt = AB(T - T_c) - C(T_c - T_{ci}) \quad (2)$$

with the initial condition

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

Motion in the phase plane $T - T_c$ can be then described by equation which is obtained by dividing relations (1) and (2). In pseudostationary state of the reactor there holds

$$dT/dt = 0, \quad \text{and} \quad dT_c/dt = 0.$$

The calculation procedure of the highest pseudostationary reaction temperature at the start (for zero degree of conversion) and during control (for degrees of conversion determined by an analysis) can be then summarised into the following points:

1) Coordinates of pseudostationary states of the reactor (Fig. 1) are determined in the phase plane by numerical solution of Eqs (1) and (2) by the method of interval halving. The calculated highest pseudostationary reaction temperature corresponding to the given degree of conversion and maximum cooling rate (*i.e.* the lowest inlet temperature and highest flow rate of heat carrier) is corrected by use of the given safety coefficients to the static, dynamic and information safeties of the reactor operation.

2) Limiting of the highest allowable pseudostationary reaction temperature by static safety results from the given set value of the static safety coefficient

$$S_{st} = r_{c,max}/r_h = (A(T_s - T_{cs}) + A_z(T_s - T_z))/(T_{ad}r_s/c_{A0}). \quad (3)$$

The highest allowable reaction temperature corresponding to the given safety coefficient is then determined by numerical solution of Eq. (3) by the method of interval halving.

3) Limitation of the highest allowable pseudostationary reaction temperature by dynamic safety results from the set value of the dynamic safety coefficient. This is defined as the ratio of critical temperature rise to instant observed temperature rise at the given temperature and degree of conversion

$$S_{dyn} = (dT/dt)_{max}/(dT/dt)_{obs} \quad (4)$$

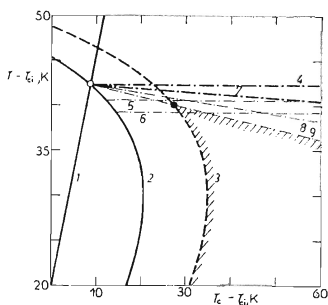
In the case that noise is significant in measurements of reaction temperature the highest allowable pseudostationary reaction temperature is limited by the information safety. The noise in temperature measurement renders difficult first of all determination of time derivative of temperature. Uncertainty in determination of derivative is described by the fixed width of noise band of derivative $(dT/dt)_n$. Coefficient of information safety is defined as the ratio of critical temperature rise to width of noise band of derivative at the given temperature and conversion degree

$$S_{inf} = (dT/dt)_{max}/(dT/dt)_n \quad (5)$$

For the control to be safe, the noise in determination of the derivative must be smaller than the critical temperature rise. The limiting value corresponds to the case

FIG. 1

Determination of highest safe pseudo-stationary temperature of reaction mixture in the phase plane for conversion degree equal to zero. $A = 1.767 \cdot 10^{-4}$, $B = 8.5$, $C = 5 \cdot 10^{-3}$; 1 pseudostationary states of the cooler, 2 pseudostationary states of the reaction mixture, 3 pseudostationary states of the mixture for $(dT/dt)_n = 0.0025$, 4 $S_{st} = 1$, 5 1.1, 6 1.2, 7 $S_{dyn} = 1$ separatrix, 8, 3, 9 5. By empty points are denoted pseudostationary states of the reactor, by full points examples of the highest safe mixture temperature for $S_{st} = 1.1$, $S_{dyn} = 5$ and $(dT/dt)_n = 0.0025$. Hatched area denotes the corresponding region of phase plane for safe operation of the reactor



when $S_{inf} = 1$, i.e. when it is just possible to recognise in the noise that the temperature rise has reached the critical value.

In both cases determination of the highest allowable pseudostationary reaction temperature requires calculation of the critical temperature rise for the instant reaction temperature and degree of conversion. This quantity is determined by numerical integration of the equation describing behaviour of the reactor in the phase plane. This equation is obtained by dividing Eqs (1) and (2). Integration is performed with the negative integration step -0.05 K and with initial conditions situated into the „close vicinity” of pseudostationary state of the reactor ($T_{cs} + 0.8 - T_{ci}$) and ($T_s - 0.2 - T_{ci}$). The Euler method has been used for integration. Integration terminates when the condition is satisfied

$$(dT/dt)_{max} \geq (dT/dt)_{obs} S_{dyn} \quad (6)$$

or

$$(dT/dt)_{max} \geq (dT/dt)_n S_{dyn} . \quad (7)$$

4) On basis of comparison of reaction temperatures corresponding to the given coefficients of static and dynamic (informative) safeties is chosen the temperature of the mixture to which it is necessary to heat or cool the reaction mixture. Chosen is of course the lower temperature.

Algorithm of temperature control consists of following steps:

1. At the start of control are given the safety coefficients, width of noise band of time derivatives of reaction temperature and parameters of the mathematical reactor model, i.e. the functional dependence of the rate of heat liberated by reaction $T_{ad}r/c_{A0}$ and parameters $A, A_z, B, C, T_{ch}, T_{ci}$ and T_z . The initial reaction temperature is chosen $T_0 - T_{c0}$ at which it is possible to arrange for the safe reactor operation. It is assumed, that at the control start two heat carriers can be used, heating carrier at the inlet temperature T_{ch} and cooling carrier at the temperature T_{ci} . To the fastest start of control then corresponds heating of the mixture on the trajectory with the fixed value of coefficients of static safety for the initial zero degree of conversion. The corresponding reaction temperature is determined by the described procedure. Calculation of this temperature at the control start is made more difficult by the fact that the quantity $(dT/dt)_{obs}$ is not known after termination of heating of the mixture. Neither is exactly known the temperature of heat carrier in the cooler after termination of heating. Determination of the highest pseudostationary reaction temperature for the given coefficient of dynamic safety has thus an iterative character. Temperature of the heat carrier in the cooler is estimated from Eq. (2), quantity $(dT/dt)_{obs}$ from Eq. (1). The iterative calculation is terminated in the case when temperature in the cooler just calculated differs in the absolute value from the value calculated in the preceding iteration by 0.05 K.

II. During the heating of mixture to the calculated value of the highest pseudostationary mixture temperature and on basis of measurements of the reaction temperature of heat carrier in the cooler and evaluation of time derivative of reaction temperature is rendered the accurate value of parameter A by use of the method of least square standard deviations and of the orthogonal transformation^{2,3}. The calculated value is used also in the following course of control for determination of the highest pseudostationary reaction temperature.

III. After termination of heating of the mixture and reaching of the calculated temperature, for the time r_d is switched on cooling of the reactor, *i.e.* used heat carrier with a low inlet temperature t_{ci} . During this time is taken sample of the reaction mixture for analysis and is determined the degree of conversion of the initial reactant.

IV. Degree of conversion is used for calculation of the new highest pseudostationary reaction temperature which corresponds to the given safety coefficients. Calculation procedure is similar to the case of the start of control. But in the given case is known the instant value of time derivative of reaction temperature $(dT/dt)_{obs}$. On basis of comparison of reaction temperatures corresponding to the given safety coefficients is then selected the temperature of the mixture in the reactor which will be the direct aim of control. In the case the calculated reaction temperature is lower than the momentous temperature observed in the reactor, the mixture is being cooled with the cooling switched on, to the value obtained by calculation. In the case it is higher, the cooling is switched off. As the reactor operates together with the cooler at switched-off position (flow rate of heat carrier into the cooler) practically in autothermal regime, the mixture in the reactor is heated by the reaction heat to the required temperature.

V. At the moment when the temperature of the mixture in the reactor has reached the calculated value the cooling is switched on or continues on for the time t_d . The whole procedure of determination of the highest allowable pseudostationary mixture temperature is then repeated. Control is terminated after reaching the degree of conversion equal to 0.9.

EXPERIMENTAL

As the model reaction was used a strongly exothermic reaction of oxidation of ethylalcohol by hydrogen peroxide, catalyzed homogeneously by iron ions. It has been found that at the used reaction conditions the reaction has an autocatalytic character⁴. Dependence of the rate of liberated heat on conversion degree of hydrogen peroxide and temperature can be described by relations ($T_{ad} = 130$ K)

$$T_{ad}r/c_{A0} = 9 \cdot 10^{11}(1 - 1.6296x) \exp(-10\,817.3/T) \quad (8)$$

in the range of conversions from 0 to 0.15, and by relation

$$T_{\text{ad}}/c_{\text{A}0} = 4.6923 \cdot 10^{12} (1-x)^{1.87} x^{0.85} \exp(-10\,817.3/T) \quad (9)$$

for higher degree of conversion. In all experiments the initial concentration of hydrogen peroxide was 2 kmol/m^{-3} , of ethanole $1.004 \text{ kmol m}^{-3}$ and of catalyst $0.001 \text{ kmol m}^{-3}$.

The measurements were performed in the stirred batch reactor with heat exchange between the reaction mixture and heat carrier in the stirred cooler. The reactor had a volume $1.7 \cdot 10^{-2} \text{ m}^3$ and was insulated thermally to environment. Heat losses into surrounding can be characterised by the value of parameter $A_z = 5 \cdot 10^{-6} \text{ s}^{-1}$. The cooling system of the reactor formed a closed loop. It consisted of the cooler with volume $2.2 \cdot 10^{-3} \text{ m}^3$ (value of parameter $B = 8.5$ and parameter $A = 1.5 \cdot 10^{-4} - 1.916 \cdot 10^{-4} \text{ s}^{-1}$) heat exchanger for bringing the temperature of the heat carrier to the inlet temperature into the cooler $T_{\text{ei}} = 289 \text{ K}$ at cooling and $T_{\text{ch}} = 343 \text{ K}$ at heating of the mixture and circulation pump (value of parameter $C = 4 \cdot 10^{-3} - 7 \cdot 10^{-3} \text{ s}^{-1}$). The experimental unit is described in detail in the previous study⁴.

For the manipulated variable for temperature control of the mixture was used the flow rate of heat carrier into the cooler (parameter C). The manipulated variable was controlled by the two-position control system switched on-off, by use of the digital measuring center Hewlett-Packard 3052A. A part of the center was the digital voltmeter 3455A and channel selector 3495A for recording the measured data in the given sampling interval and switching on and off the circulating pump of the cooling reactor system, of the control unit and digital computer Hewlett-Packard 9835A. The attached digital time unit enabled the control in real time.

In experiments which started by charging the catalyst solutions continuously were measured the reaction temperature in the reactor, temperature of the heat carrier in the cooler and at the inlet into the cooler by use of a thermocouple Fe-constantan with time constants 2–3 s on the sampling interval 10 s. Measured values were filtered in two stages⁵ by use of the digital exponential filter and by smoothing on the interval 240 s by use of the third degree polynomial. Constants of the polynomial were evaluated by the linear method of multiple regression. From the measured reaction temperature were smoothed after filtration the time derivatives of reaction temperature by use of the three point difference chart. From 100 measured and filtered reaction temperatures was also simultaneously determined the standard deviation of noise.

Analysis. During the experiment samples of the reaction mixture were taken periodically. Manganometrically was determined the hydrogen peroxide content in the mixture and degree of conversion was evaluated. Degrees of conversion were fed through the keyboard to the digital computer.

RESULTS AND DISCUSSION

The aim of temperature control of the reaction mixture was to keep the batch reactor with exothermic reaction in pseudostationary state which was unstable at the open control loop. An example of calculation of coordinates of such state in the phase plane is given in Fig. 1 for parameters of the system, which correspond to the experiments.

Effect of cooler inertia. Basis for preserving the pseudostationary state by two-position control is the alternate motion of temperature up and down in vicinity of the

state. From dynamic point of view the control of temperature is safe when it is possible to arrange that, after switching to the highest cooling rate, the temperature response of the mixture does not exceed the region of static safety. The rate of this response depends on inertia of the cooler, *i.e.* how quickly the temperature of the heat carrier in the cooler T_c (manipulated state variable) reacts to changes in flow rate of the heat carrier into the cooler (parameter C). Inertia of the cooler has an asymmetric character at two-position control. After switching off the flow rate of heat carrier the effect of this inertia is determined by the heat transfer coefficient and by thermal capacity of the cooler (parameters A and B). It cannot be changed for the given reactor design. Inertia can be affected after switching on the flow rate of the heat carrier by the flow rate (parameter C). The studied system can be characterised as the system with significant inertia effects of the cooler on the course of control of reaction temperature. This is demonstrated by the results of experiments (Fig. 2) when the effect of flow rate of the heat carrier on control safety is studied, *i.e.* the effect of parameter C of the reactor model. The value of parameter $C = 4 \cdot 10^{-3} \text{ s}^{-1}$ can be considered limiting when still for the given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 3$ the control safety of reaction temperature is preserved. The results of experiments confirm the conclusions resulting from the simulation calculations made in the previous study¹. In cases when the cooler has a high inertia it is necessary to fix the coefficient of dynamic safety within the range from 3 to 5 for control of reaction temperature by use of the proposed control algorithm.

Effect of uncertainties in information on the system. At the control of mixture temperature by use of algorithm control based on the mathematical model of the system the control safety is related with reliability of informations on the system. It is suitable to divide these informations into several groups.

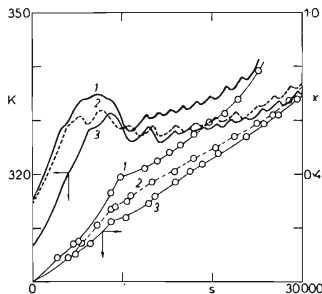


FIG. 2

Control of reaction temperature on given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 3$. Effect of parameter C . $t_d = 300$, $(dT/dt)_n = 0.0025$; 1 $C = 4 \cdot 10^{-3}$, 2 $5 \cdot 10^{-3}$, 3 $6.273 \cdot 10^{-3}$. Empty points denote degrees of conversion of the reactant x determined by analysis of samples of the mixture

The studied control algorithm is an example of forward nonadaptive control. Safety control thus necessarily depends on reliability of data fed into the reactor model, *i.e.* on accuracy of fixed parameters. How difficult is the feedback temperature control of the mixture in the batch reactor with exothermic reaction depends first of all on dependence of requirements of the cooling rate on the degree of conversion. With reactions of positive order they are greatest at the beginning of reaction, with autocatalytic reactions in the region of maximum of the mixture reactivity. In the given case this is in the region close to the conversion degree 0.2 to 0.3. Of basic significance is thus reliability of the given functional dependence, which describes the effect of conversion degree on reaction rate. Directly related is the effect of changes of certain non-measured parameters. For example the effect of changes in catalyst concentration, reaction initiator and scaling of heat transfer areas.

The effect of these parameters has been studied by use of concentration changes

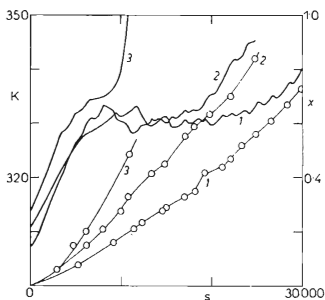


FIG. 3

Control of reaction temperature on given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 5$. Effect of catalyst concentration. $C = 5 \cdot 10^{-3}$; 1 $c_{cat} = 0.001$, 2 0.0011 , 3 0.0012 . Other symbols used are the same as those in Fig. 2

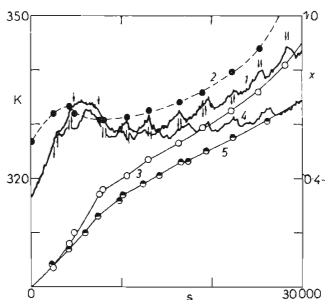


FIG. 4

Effect of noise on control of reaction temperature on given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 5$. $\sigma = 0.5$; 1 reaction temperature after filtration, $(dT/dt)_n = 0.0025$, 2 highest safe pseudostationary reaction temperature calculated for the given conversion degree by use of the mathematical model, 3 conversion degree x , 4 reaction temperature after filtration, $(dT/dt) = 0.055$, 5 conversion degree x . Arrowheads denote switching on (\uparrow) and off (\downarrow) of flow rate of heat carrier into the cooler. Other symbols used are the same as those in Fig. 3

of the catalyst which is at the beginning of the reaction charged into the mixture and by use of correction of the functional dependence $T_{ad}r/c_{A0}$ describing the effect of conversion degree on rate of heat liberated by reaction. Results of these experiments are given in Figs 3 and 5. In Fig. 3 are given results of experiments at which was step by step increased the catalyst concentration by 10%. In Fig. 5 (curve 2), the mixture reactivity was recalculated by use of the model corrected so that the calculated value was by 10% lower than in the standard state (Fig. 1, curve 2). Results of experiments point to the fact that it is necessary to take into consideration the high parametric sensitivity of safety coefficients. But they simultaneously confirm that the studied control algorithm is sufficiently "robust" to changes in reactivity for 10 relative %, which are in actual situations usual.

The highest safe reaction temperature in systems with the effect of noise is limited by noise by which are affected the measured data. In the given case these are reaction temperatures in the reactor and from them evaluated time derivatives. The experience demonstrates that the noise of the measured mixture temperature significantly affects just these control algorithms which are based on evaluation of time derivatives of the measured temperature. Under conditions of laboratory verification of the

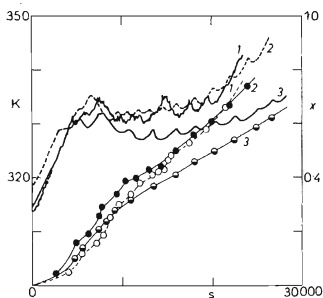


FIG. 5

Control of reaction temperature on given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 3$. The effect of noise and inaccuracies of mathematical model. 1 Effect of additive noise of reaction temperatures, $\sigma = 0.5$, $(dT/dt)_n = 0.005$, 2 effect of inaccuracies of the model, $(dT/dt)_n = 0.0025$, 3 control without noise, $(dT/dt)_n = 0.0025$. Other symbols used are the same as those in Fig. 3

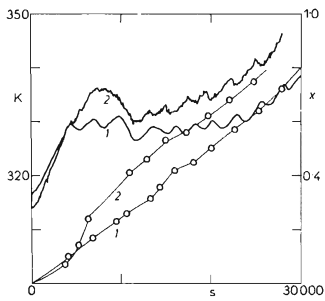


FIG. 6

Control of reaction temperature on given safety coefficients $S_{st} = 1.1$ and $S_{dyn} = 3$. The effects of noise and of time needed for analysis of the mixture. $(dT/dt) = 0.0025$; 1 effect of time for analysis of mixture, $t_d = 600$, 2 effect of additive noise of reaction temperature, $\sigma = 0.5$, $t_d = 300$. Other symbols used are the same as those in Fig. 3

control algorithm the effect of noise on control has been negligible. The magnitude of noise can be characterised by standard deviation $\sigma = 0.01$ to 0.02 K, and width of noise band of time derivative of temperature by the value $(dT/dt)_n = 0.0025$ K \cdot s^{-1} . The estimate of this quantity is perhaps affected by the largest uncertainty. For calculation was used the approximate relation

$$(dT/dt)_n = 2\sigma/\Delta t, \quad (10)$$

where Δt denoted the sampling interval of temperatures of the reaction mixture.

The effect of noise on control safety of mixture temperature was thus studied by use of additive „white” noise generated by the digital computer. In all experiments the noise was generated that could be characterized by standard deviation $\sigma = 0.5$ K. The control safety of mixture temperature then depends directly on efficiency of the used filtration method of measured mixture temperatures in the reactor. Results of experiments studying the effect of noise are presented in Figs 4 to 6.

In some of experiments (Fig. 4, curve 1, Fig. 6, curve 2) was assumed that the width of noise band of derivative $(dT/dt)_n$ after filtration could be given as $(dT/dt)_n = 0.0025$ K s^{-1} . This means that data filtration is completely effective. As demonstrated the results of experiments this assumption proved insignificant. The „residual noise” after filtration significantly worsened the quality of control and reduced its safety. It was demonstrated, that magnitude of this noise might be characterised by width of noise band of time derivative of reaction mixture $(dT/dt)_n = 0.005$ K s^{-1} . When this estimated value was used for determination of the highest safe temperature of the reaction mixture (Fig. 4, curve 3, Fig. 5, curve 1) control of reaction temperature by use of the proposed control algorithm was safe.

CONCLUSION

Results of experimental verification of control algorithm of the batch reactor at the highest safe pseudostationary temperature of reaction mixture proved that the proposed control algorithm was sufficiently „robust” to changes in mixture reactivity and to changes of measured system parameters by 10% relative. Basic significance for control safety has the efficiency of filtration of measured reaction temperature and reliability of the estimate of the band width of time derivative of reaction temperature.

LIST OF SYMBOLS

$A = k_h P / (V_c \rho_c)$	parameter characterising heat transfer between reactor and cooler (s^{-1})
$B = V_c \rho_c / (V_c c_{pe} \rho_c)$	ratio of thermal capacities of reaction mixture in reactor and heat carrier in the cooler
$C = F_c / V_c$	reciprocal value of residence time of heat carrier in the cooler (s^{-1})

c_{A0}	initial concentration of reactant (kmol m^{-3})
c_{cat}	catalyst concentration (kmol m^{-3})
c_p, c_{pc}	specific heat of mixture and heat carrier ($\text{kJ kg}^{-1} \text{K}^{-1}$)
F_c	volumetric flow rate of heat carrier into the cooler ($\text{m}^3 \text{s}^{-1}$)
k_h	heat transfer coefficient ($\text{kW m}^{-2} \text{K}^{-1}$)
P	heat transfer area (m^2)
R	gas law constant
r	reaction rate ($\text{kmol m}^{-3} \text{s}^{-1}$)
r_c, r_h	cooling rate and rate of heat liberated by reaction (K s^{-1})
T	reaction (mixture) temperature (K)
T_{ad}	adiabatic temperature rise (K)
T_c	temperature of heat carrier in cooler (K)
T_{ci}	temperature of heat carrier at the inlet into the cooler at cooling of the mixture (K)
T_{ch}	inlet temperature of the heat carrier at heating of the mixture (K)
t	reaction time (s)
t_d	time for analysis of the mixture for determination of conversion degree (s)
S	safety coefficient
V, V_c	volume of reactor and cooler (m^3)
x	degree of conversion of reactant
ρ, ρ_c	specific density of mixture and heat carrier (kg m^{-3})
σ	standard deviation (K)

Subscripts

max	largest value of quantity
obs	observed value of quantity
o	initial value of quantity
s	value in pseudostationary state
z	vicinity of reactor

REFERENCES

1. Horák J., Jiráček F.: This Journal 48, 711 (1983).
2. Eykhoff P.: System Identification. Wiley, London 1974.
3. Soukup J.: System Identification, ČVUT textbook (in Czech), Prague 1982.
4. Jiráček F., Horák J.: This Journal 48, 449 (1983).
5. Weber R.: AIChE J. 26, 142 (1980).

Translated by M. Rylek.