ADAPTIVE NONISOTHERMAL COMPUTER CONTROL OF A BATCH REACTOR

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The adaptive temperature control method of reaction mixture in a batch reactor with strongly exothermic reaction is verified experimentally. The utility degree of cooling capacity of the reactor is kept on the required value by use of a two-position controller which is changing the flow rate of heat carrier into the cooler. The method is based on continuous identification of properties of the system in actual time by use of the Hewlett-Packard 3 052 A computer centre. During the experiments temperature of mixture in the reactor and temperature of heat carrier in the cooler were measured. From the obtained data and their time derivatives reactivity of the mixture, heat transfer coefficient and utility degree of cooling capacity of the reactor were evaluated. In the experiments were studied the effects of cooler inertia and of additive noise in the temperature measurement on safety and quality of control. Experimental results have proved that the method is applicable to control of reaction temperature even under conditions when properties of the system are changing significantly *e.g.* reactivity of the mixture and dynamic properties of the manipulated variable and when pseudostationary states of the reactor are unstable in the open control loop. They simultaneously point to a significant effect of noise on system identification.

Among the first studies where an analysis of adaptive control of a batch reactor was performed belong studies by Aris and coworkes[1,2]. From recent studies, it is possible to mention the paper by Horn[3]. For identification of system properties these studies use first of all the fact, that at the beginning of the process, when the mixture is heated to the reaction temperature it is possible to determine the actual value of the heat transfer coefficient from the temperature changes. This value is then in the region, when the chemical reaction is already under way, used for determination of reaction rate from the heat balance of reaction mixture. This evaluation enables to propose the optimal moment for switching the reactor system into operation and further to propose parameters of the feedback controller for keeping temperature of the mixture at the constant — required value.

In the study by Horák, Jiráček and Ježová[4] a simple adaptive method of reaction temperature control has been proposed, where the heat transfer rate is altered by use of a two-position controller. The utility degree of cooling capacity of the reactor may be kept on the given value by this method. The utility degree is evaluated from ratio of time at which the cooling system of the reactor is in operation to time for which it is out of operation. The method is applied to reactors with small inertia of the cooling system. In the study are experimentally verified results of simulation on a mathematical model in the laboratory batch reactor in which activity of the catalyst is changing. In the study by Jiráček and Horák[5] the considered control method is studied on the case of a reactor with large inertia of the cooling system. The results of mathematical simulations have proved a significant effect of dynamic properties of the cooling system on reaction temperature control.

In this study have been verified experimentally the effects of inertia of the reactor cooling system, of noise in information outputs on adaptive reaction temperature control under conditions when for control is used direct digital control (on line computer control in real time).

THEORETICAL

Mathematical Model of the System

Description of dynamic behaviour of the system is based on the material and heat balance in the perfectly stirred batch reactor

$$\mathrm{d}x/\mathrm{d}t = r/c_{\mathrm{Ao}} \tag{1}$$

$$(1/A)(\mathrm{d}T/\mathrm{d}t) + T = T_{\mathrm{c}} + rT_{\mathrm{ad}}/(Ac_{\mathrm{Ao}}) \tag{2}$$

$$\left(\frac{1}{AB + C}\right)(\mathrm{d}T_{\mathrm{c}}/\mathrm{d}t) + T_{\mathrm{c}} = \frac{ABT + CT_{\mathrm{ci}}}{AB + C} \tag{3}$$

with initial conditions

$$t = 0\,; \quad x = 0\,; \quad T = T_0\,; \quad T_{\mathrm{c}} = T_{\mathrm{co}}\,.$$

In pseudostationary states of the reactor (x is supposed to be constant) there holds

$$\mathrm{d}T/\mathrm{d}t = 0; \quad \mathrm{d}T_{\mathrm{c}}/\mathrm{d}t = 0\,. \tag{4}$$

Adaptive Control Method

Theoretical value of the utility degree of cooling capacity of the reactor (further on called only utility degree) is defined as the ratio of rate of heat generated by reaction to the maximum possible cooling rate at the given temperature of the mixture

$$R_{\mathrm{s}} = r_{\mathrm{h}}/r_{\mathrm{c}} = (rT_{\mathrm{ad}}/c_{\mathrm{Ao}})/(A(T_{\mathrm{s}} - T_{\mathrm{c}}))\,. \tag{5}$$

To start up the adaptive control an estimate of the reaction temperature for the first switching on of the cooling system has to be known. At the temperature of "the first evaluation" of the utility degree the reactor regime must be safe.

Temperature of the mixture in the reactor is controlled by two position variation of the cooling rate, closing and opening of passage for the heat carrier into the cooler.

The cooling rate during one cycle thus becomes equal to zero or to the largest value. Switching over of cooling is governed by conditions for the algorithm "relay with hysteresis".

The momentous utility degree is determined from temperature response of the mixture on switching on or off of cooling, given by

$$R_e = t_c/(t_a + t_c)\,. \tag{6}$$

From the utility degree R_e is determined by extrapolation the new value of mixture temperature T_s, at which R_e is reaching just the required value. Determination of the mixture temperature by extrapolation is based on the assumption that reaction rate depends only on temperature according to the Arrhenius equation and that cooling rate is proportional to temperature difference between that of the mixture and inlet temperature of the heat carrier entering the cooler, which is given by relations

$$r_h = C_1 \exp\left[E(T_s - T_{ci})/RT_sT_{ci}\right] \tag{7}$$

$$r_c = C_2(T_s - T_{ci})\,. \tag{8}$$

For the switching over cooling cycle the relation

$$R_e = C_{ex} \exp\left[E(T_s - T_{ci})/RT_sT_{ci}\right](T_s - T_{ci})^{-1} \tag{9}$$

holds approximately where $C_{ex} = C_1/C_2$ is a constant including variable parameters of the system.

The constant C_{ex} can be continuously evaluated from the momentous value of utility degree R_e. After substitution of this constant into Eq. (9) together with the required utility degree it is then possible to determine by numerical solution of the equation (the method of halving intervals is used) temperature of the mixture T_s at which the utility degree is equal to the given value. By succesive repeating of experimental determination of the utility degree R_e and successive selection of mixture temperature T_s is the utility degree kept on required constant value.

Algorithm of "Relay With Hysteresis"

Switching over of the reactor cooling is governed by following conditions. Cooling of the reactor is switched on, when

$$T \geqq T_s + T_{reg}\,. \tag{10}$$

Cooling of the reactor is switched off when

$$T \leqq T_s - T_{reg} . \tag{11}$$

Inside the interval $(T_s - T_{reg}; T_s + T_{reg})$ the previous state of the controller is held.

Modified Adaptive Control Method

Similarly as with the preceding method is assumed that the activation energy of reaction is known and during the control the mixture temperature in the reactor is measured. For the rate of heat generation the relation holds

$$r_h = C_1 \exp(-E/\boldsymbol{R}T) , \tag{12}$$

where the constant C_1 is characterising reactivity of the mixture, *e.g.* effect of conversion of the key component and catalyst activity on reaction rate.

Unlike the last method, it is assumed that is also measured temperature of the heat carrier in the cooler. Cooling rate is proportional to temperature difference between that of mixture in the reactor and temperature of heat carrier in the cooler, which is given by

$$r_c = C_2(T - T_c) . \tag{13}$$

The constant C_2 is identical with the parameter A in Eq. (*2*).

The modified control method is based on successive evaluation of each of constants C_1 and C_2 on the given identification interval. On basis of the measured temperatures of mixture and heat carrier and their derivatives per time it is possible to determine at the switched-on cooling of the reactor, the constant C_2 from relation (Eq. (*3*))

$$C_2 = \frac{\mathrm{d}T_c/\mathrm{d}t + C(T_c - T_{ci})}{B(T - T_c)} \tag{14}$$

and at the switched-on or off reactor cooling the constant C_1 from relation (Eq. (*2*))

$$C_1 = \frac{\mathrm{d}T/\mathrm{d}t + C_2(T - T_c)}{\exp(-E/\boldsymbol{R}T)} . \tag{15}$$

At the end of the identification interval are then determined the average values from the calculated constants. It is possible to obtain the initial estimate of constant C_2 *e.g.* from measurements of the heat transfer intensity in the reactor without chemical reaction by evaluation of heating period of the mixture to the in advance fixed temperature.

After completing each identification period of the system the constants are used for extrapolation of the reaction temperature T_s corresponding to the required utility degree of cooling capacity of the reactor R_c. Extrapolation is based on numerical solution of the relation

$$R_c C_1 \exp\left(-E/RT_s\right) - C_2(T_s - T_c) = 0 \qquad (16)$$

which results from the definition equation (5).

As the first estimate of temperature of the carrier in the cooler is substitued the inlet temperature of heat carrier T_{ci}. After calculation of the extrapolated mixture temperature the extrapolation procedure is repeated with the temperature of the heat carrier estimated from Eq. (3) for the case of pseudostationary state of the reactor

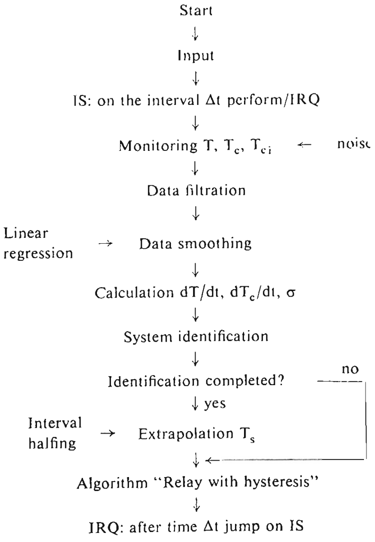

FIG. 1
Block diagram of computer program

according to

$$T_c = \frac{C_2BT_s + CT_{ci}}{C + C_2B} . \qquad (17)$$

On basis of the algorithm "relay with hysteresis" it is then decided whether the reactor cooling should be switched-off or on and the whole procedure is repeated. Simplified block diagram of the computer program is given in Fig. 1.

EXPERIMENTAL

Model Reaction

For model reaction was used a strongly exothermic oxidation of ethanol with hydrogen oxide in liquid phase, catalysed homogeneously by ferric ions[4]. The results of evaluation of mixture reactivity at experimental verification of the modified adaptive control method demonstrated that under the used reaction conditions (Table I) the reaction was autocatalytic (Fig. 2). For description of dependence of reactivity constant C_1 on degree of conversion of hydrogen peroxide it is possible to use relation

$$C_1 = 4.6923 \cdot 10^{12}(1 - x)^{1.87}\, x^{0.85} . \qquad (18)$$

TABLE I

Parameters of chemical reaction and of laboratory apparatus

Parameter	Value	Dimension
E	90	kJ mol^{-1}
T_{ad}	130	K
Initial concentration of H_2O_2	1·024	kmol m^{-3}
Initial concentration of C_2H_5OH	2·0	kmol m^{-3}
Catalyst concentration	0·001	kmol m^{-3}
V	0 017	m^3
V_c	0·0022	m^3
P_c	0·068	m^2
F_c	1·052 . 10^{-5}	m^3 s^{-1}
A	1·5 . 10^{-4}	s^{-1}
B	7·72	
C	4·78 . 10^{-3}	s^{-1}
T_{ci}	281—283	K
T_{reg}	0·5	K
Δt	5	s
Interval of data smoothing	60—120	s
Identification interval	300—600	s

Decrease of reactivity at the beginning of reaction after feeding the catalyst solution into the mixture is perhaps related with the content of oxidation products in the mixture which may activate the catalyst. All experiments were performed at the same initial composition of the reaction mixture and catalyst concentrations.

Laboratory Reactor

The measurements were performed in the batch reactor with heat exchange between the reaction mixture and heat carrier in a stirred cooler (Fig. 3). The reactor was a polyethylene vessel, insulated thermally with foamy polystyrene from environment. The rate of the heat transfer with environment was 25 times smaller than the heat transfer rate into the cooler. The cooler was formed of glass vessel whose internal surface was covered by foamy polyethylene. By its thickness could be fixed the heat transfer coefficient. The cooling system of the reactor formed a closed loop. Circulation of the heat carrier in the loop was arranged for by use of a peristaltic pump. Its operation was actuated by two-position system open-closed, operated by computer. Parameters of the laboratory apparatus are given in Table I.

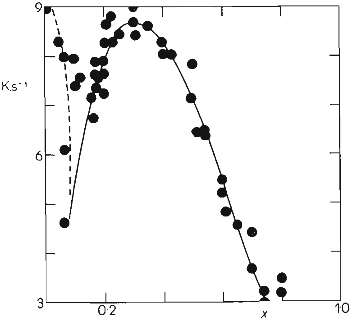

FIG. 2

Measured mixture reactivity in dependence on degree of conversion of hydrogen peroxide. Solid line denotes the calculated dependence

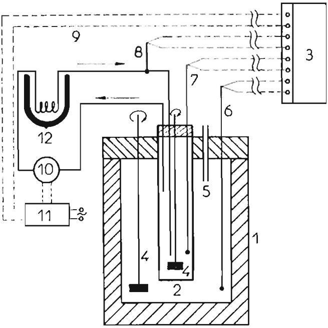

FIG. 3

Laboratory apparatus. *1* Reactor with thermal insolation, *2* cooler, *3* measuring unit, *4* mixer, *5* feeding of mixture and catalyst, sampling of mixture; *6* thermocouple for temperature measurements in reactor; *7* thermocouple for temperature measurements of heat carrier in cooler; *8* thermocouple for temperature measurement at inlet of heat carrier; *9* connection of circulation pump to measuring centre; *10* circulating pump; *11* relay; *12* cooling of heat carrier

During the experiment were automatically measured in the given sampling interval[7], temperature of mixture in the reactor by iron-constantant thermocouples, temperature of the heat carrier in the cooler and at its inlet into the cooler. Experimentally determined time constants of thermocouples were 2 to 3 s. Temperature was measured with the accuracy ±0.05 K. While the reaction took place, the reaction mixture was sampled, for determination of conversion of hydrogen peroxide. Starting of the experiments was similar in all experiments. Reaction mixture without the catalyst was heated to initial temperature 326 to 327 K, initial temperature of heat carrier in the cooler was 313 K. In some experiments the effect of initial mixture temperature on control was studied. During heating of the mixture to the initial temperature the heat transfer coefficient was evaulated (constant C_2).

Digital Measuring Unit

For control of the reactor in real time the "Automatic Data Acquisition System 3 052" by Hewlett--Packard was used. The unit consisted of the numerical voltmeter 3 455 A, channel switch 3 495 A, numerical time unit 98 035 A and control system unit, bench calculator Hewlett-Packard 9 835 (64 Kbites of the operating memory).

Data Filtration

In some experiments the additive "white noise" was generated on the automatic computer of the measuring centre. The generated pseudorandom numbers with constant probability distribution with values in the range from 0 to 1 were transformed to random numbers with the normal probability distribution, characterized by the standard deviation σ and zero mean value. The values of noise determined in this way were in each sampling interval added to measured temperatures of the mixture and of the heat carrier.

Data filtration took place in two stages. In the first one the nonlinear exponential filter, proposed in the study by Weber[6] was used for data filtration. Filtration was performed according to the relation

$$T_n - T_{n-1} = f(\Delta X_n)\,\Delta X_n \tag{19}$$

$$f(\Delta X_n) = \text{Min}\left(1, \left|\frac{\Delta X_n}{K\sigma}\right|\right) \tag{20}$$

T_n and T_{n-1} are filtered values of the measured quantity in the sampling interval Δt_n and Δt_{n-1}. ΔX_n difference between the momentous value of measured quantity with noise and value which was filtered last. In the second stage the data were filtered *i.e.* dependence of measured quantities on the reaction time was smoothed by use of the third degree polynomial. The constants of polynomial were evaluated by the method of multifold linear regression. After calculation of constants the value of quantity at the end of the smooting interval was calculated by use of the polynomial and the interval was shifted by the given time length. The whole data filtration procedure was then repeated. From the calculated values are then determined their time derivatives by use of the three-point difference scheme.

Standard deviation of the additive noise has been always evaluated continuously during the control from 100 temperature measurements. Noise in the given sampling interval was estimated as the difference of the measured temperature with noise and value calculated after filtration by use of the polynomial. Calculated standard deviations were in a good agreement with the value given at generation of noise (maximum deviation ±0.1).

RESULTS AND DISCUSSION

In cases when temperature of the mixture in the batch reactor is not a fixed quantity it could be kept at the highest value, under which the safety of operation is still secured. Keeping of temperature at the highest admissible value corresponding to the given utility degree of cooling capacity leads to the maximum reactor output[4]. This method of operation of the batch reactor can be easily realised by adaptive control method only. During the reaction, due to changing conversion of the mixture, properties of the system in an actual reactor significantly change and moreover, the effect of nonidealities of the system and of dynamic properties of the manipulated variable can appear.

One of basic problems of adaptive temperature control is thus collection of information on momentous state of the system, *i.e.* identification of the system[8]. In this study was studied the effect of inertia of the reactor cooler and effect of noise in information outputs on control under conditions, when in the batch reactor a strongly exothermic autocatalytic reaction takes place and pseudostationary states of the reactor are unstable in the open control loop. This assumption is verified by calculation of the heat generated by reaction and of the cooling rate for system parameters, which were evaluated from experiments (Fig. 4).

Adaptive Control Method

At the proposed simple temperature control method is assumed that the safe reaction temperature for the first identification of the system is known. Up to the moment of the first evaluation of the utility degree there is no information on the state of the system. The risk of uncontrolled temperature increase of the mixture thus depends first of all on selection of the temperature of the first identification (Figs 5 to 7). The results of experiments have proved a significant effect of cooler inertia on control[5]. Inertia of the cooler resulting from its large thermal capacity (Table I) causes that the result of switching on or off of the cooling is not observed immediately and consequently overcontrol results. With regard to asymmetric character of the cooler behaviour[5] the overcontrol values are not equal for on and off of the controller. Under conditions used in this study is especially significant the temperature overcontrol below the value given by the hysteresis band, after switching off of the cooling as the result of a high flow velocity of the heat carrier in the cooler and thus of cold accumulated in the cooler. Inertia of the cooler thus leads on one side to increase of time needed for determination of the experimental value of utility degree R_c and on the other side to the increase of the transition time to the next extrapolated mixture temperature T_s. This decreases adaptibility of control. This effect becomes disadvantageous especially when the reaction terminates, when changes of the degree of conversion have a relatively large effect (in extrapolation of temperature the effect of conversion is neglected). The cooler inertia has also negative consequences on stability of the control algorithm. The used method neglects in determination of the utility degree of cooling capacity the amount of heat accumulated in the cooler (temperature of the heat carrier in the cooler is approximated by inlet temperature of the carrier). So the obtained result is distorted and can cause an oscillation instability (Figs 5 and 7). Its result can be a failure of control *i.e.* uncontrolled increase of temperature of the mixture.

Results of experiments have proved that at conditions when the cooler has a large inertia and requirements on the cooling rate increase during the process, the inertia of the cooler is a serious danger. These negative consequences result first of all from inaccurate and delayed information on the instantaneous state of the system.

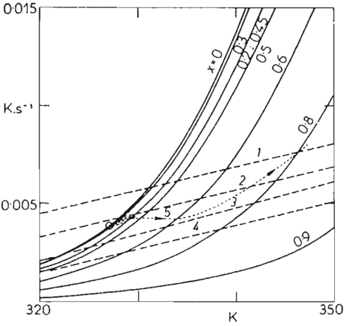

FIG. 4

Calculated rate of heat generation and cooling rate in dependence on temperature. Solid line denotes dependence of rate of heat generation, dashed line the cooling rate and narrow dashed line the optimal trajectory of mixture temperature in reactor for $R_s = 0 8$. *1* $T_c = 283$ K; *2* 293 K; *3* 303 K; *4* 308 K

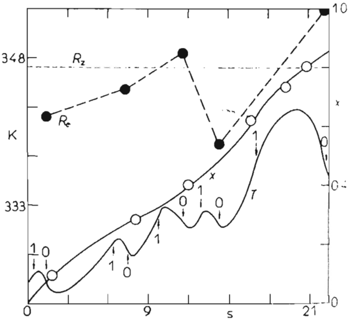

FIG. 5

Adaptive temperature control of mixture for $R_s = 0·8$. Measured temperature dependence of mixture in reactor, degree of conversion of hydrogen peroxide and utility degree of cooling capacity on reaction time. Arrows denote the moment of switching over of cooling; 0 cooling switched off, 1 cooling switched on

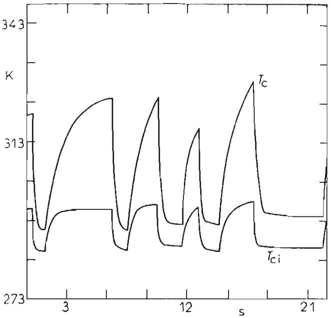

FIG. 6

Measured temperature of heat carrier in the cooler and at inlet into cooler in dependence on reaction time for the case given in Fig. 5

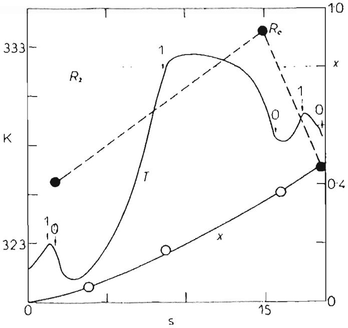

FIG. 7

Adaptive control of mixture temperature for $R_s = 0.8$. Effect of initial temperature of mixture. Symbols used are the same as in Fig. 5

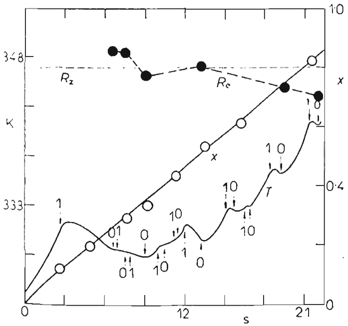

FIG. 8

Modified adaptive method control for $R_s = 0.8$. Symbols used are the same as in Fig. 5

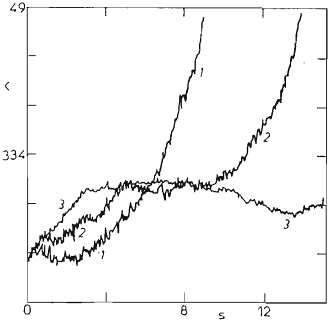

FIG. 9

Modified adaptive control method for $R_s = 0.8$. Effect of noise on mode of control. *1* Measured mixture temperature in dependence on reaction time for $\sigma = 0.5$ and interval of data smoothing 60 s; *2* $\sigma = 0.5$ and 120 s; *3* $\sigma = 0.25$ and 120 s

Modified Adaptive Control Method

The discussed adaptive control method has a number of advantages, especially simplicity and small requirements on the number of information outlets from the system. Its disadvantage is an insufficient adaptibility resulting from discontinuity of the system identification and inaccurate information on the momentous state of the cooler. In the case of direct digital control by a computer possibilities of the considered control model are also not fully used. The modified control method is based on continuous identification of the system during the whole process. On basis of temperature measurements of mixture in the reactor and heat carrier in the cooler, reactivity of the mixture and heat transfer coefficient are evaluated. Together with the known estimate of heat capacity of the cooler and flow rate of the heat carrier into the cooler (parameters *B* and *C*) basis is made for elimination of the inertia effect of the cooler on mode of control, first of all on extrapolation of temperature of the mixture corresponding to the giver utility degree of thermal capacity of the reactor. The results of experiments have proved (Fig. 8) that it is possible to decrease significantly the occurrence of the oscillation instability control by the given procedure.

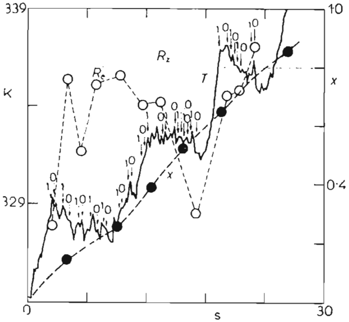

FIG. 10

Modified adaptive control method for $R_s = 0{\cdot}8$. Effect of noise on mode of control $\sigma = 0{\cdot}5$; interval of data smoothing 120 s; identification interval 300 s. $K = 5$

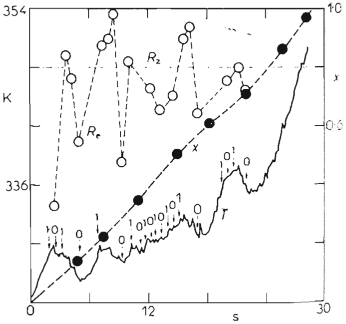

FIG. 11

Modified adaptive control method for $R_s = 0{\cdot}8$. Effect of noise on control mode. Smoothing interval 600 s; other symbols are identical with those in Fig. 10

The effect of uncertainty in the estimate of activation energy of the reaction and errors in determination of reactivity of the mixture and heat transfer coefficient of constants C_1 and C_2 on identification has been verified during the experiment. It has been found that the error in the estimate of activation energy ±10% relative, is causing an error in the estimate of the extrapolated mixture temperature T_s ± 1.5% rel. and equal error in determination of reactivity of the mixture and the heat transfer coefficient an error ±2.5% rel.

Effect of Noise on Control

In a real reactor the outlet informations of the system are always affected by noise occurring by disturbances in the controlled system and in the control circuit. From analysis of data measured in the laboratory reactor results that experimental temperatures of mixture and heat carrier are affected only by a negligible noise. The effect of noise had to be thus simulated by use of the additive noise generated in the control computer. During the experiment the effect of magnitude of noise, smoothing interval (Fig. 9) and identification interval (Figs 10 and 11) on control operation were studied. On basis of thus obtained data results that the noise represents a serious problem in application of adaptive methods for control of temperature of the mixture in the batch reactor. Consequences of the effect of noise on control operation have a similar character as the effect of cooler inertia and lead to the oscillation instability of control. The control quality then depends first of all on efficiency of data filtration which are obtained during identification of the system. The used filtration method of data appeared less efficient.

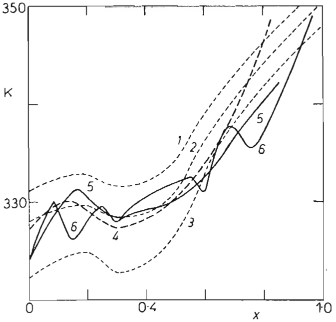

FIG. 12

Comparison of calculated and experimental dependences of mixture temperatures in pseudostationary reactor states on conversion degree for R_s = 0·8. *1* T_c = 283 K; 2 293 K; *3* 303 K; *4* optimal trajectory (curve *5* in Fig. 4); *5*, *6* measured trajectories (Figs 10 and 11)

CONCLUSIONS

In this study has been experimentally verified the adaptive method of temperature control of mixture in the reactor, based on direct digital control of the process by computer in real time. The results of experiments have proved that application of this method may be an effective way toward increase in the reactor output under conditions when properties of the system significantly change during the process and cooling system of the reactor has a large inertia. From results of experiments, which are in a very good agreement with that obtained by mathematical simulation (Fig. 12) results that quality and security of temperature control decreases with magnitude of noise by which the measured data are affected.

LIST OF SYMBOLS

$A = k_h P_c/(V c_p \varrho)$	parameter characterising intensity of heat transfer between reaction mixture in reactor and heat carrier in cooler (s^{-1})
$B = V c_p \varrho/(V_c c_{pc} \varrho_c)$	ratio of thermal capacities of reaction mixture in reactor and heat carrier in cooler
$C = F_c/V_c$	parameter characterising residence time of heat carrier in cooler (s^{-1})
C_1, C_2, C_{cx}	constants
c_{A0}	initial concentration of the key component (kmol m^{-3})
c_p, c_{pc}	specific heat of reaction mixture and heat carrier (kJ kg^{-1} K^{-1})
E	activation energy of reaction (kJ mol^{-1})
F_c	volumetric flow rate of heat carrier into the cooler (m^3 s^{-1})
K	constant in nonlinear filter
k_h	heat transfer coefficient (kW m^{-2} K^{-1})
P_c	heat exchange area of cooler for heat transfer (m^2)
R	gas constant
R_s, R_e	set value and experimental value of utility degree of cooling capacity
r_c	cooling rate relative (Ks^{-1})
r_h	rate of heat generated by reaction relative (Ks^{-1})
T	temperature of reaction mixture (K)
T_c, T_{ci}	temperature of heat carrier in cooler and at inlet into the cooler (K)
T_0, T_{co}	initial temperature of mixture in reactor and heat carrier in cooler (K)
T_{ad}	adiabatic heating of reaction (K)
T_{reg}	width of hysteresis band of controler (K)
T_s	temperature of mixture corresponding to the given utility degree of cooling capacity of reactor (K)
t	reaction time (s)
t_a, t_c	time for which the cooling is switched-off or on (s)
V, V_c	mixture volume in reactor and of heat carrier in cooler (m^3)
x	degree of conversion of key component
Δt	sampling interval (s)
σ	standard deviation of noise
ϱ, ϱ_c	density of reaction mixture and heat carrier (kg m^{-3})

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