

## NONISOTHERMAL CONTROL OF A BATCH REACTOR WITH EXOTHERMIC REACTION USING A MATHEMATICAL MODEL

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A method of nonisothermal control of a batch reactor with exothermic reaction is suggested and tested. The control algorithm is based on jump changes in the inlet temperature of coolant, which is chosen so that the highest difference between the temperature of the reaction mixture and the temperature of entering coolant is held at a value near the highest temperature difference at which the stable pseudostationary state still exists. A total model of the system is used for the prediction of the entering coolant temperature.

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A method of nonisothermal control of a reactor with exothermic reaction, based on stepwise jump changes in the inlet temperature of coolant, has been suggested and tested previously<sup>1</sup>. The entering coolant temperature is so varied that the temperature difference between the reaction mixture and the entering coolant in the temperature maximum of the reaction mixture is held at a value near the highest temperature difference at which stable pseudostationary states still exist. A zeroth order mathematical model has been employed for the prediction of the corresponding inlet temperature of coolant.

This model is convenient in that the knowledge of the activation energy is sufficient for the control while the reaction order need not be known. On the other hand, however, the change in the degree of conversion during the transition process and the inertia of the cooling system are disregarded in this model. As a result of this simplification, pessimistic predictions are obtained for reactions with positive overall orders, whereupon the cooling capacity of the reactor is not fully exploited. Actually, the change in the degree of conversion during the transition process leads here to a decrease in reactivity and, consequently, in the production of heat; and the large inertia of the cooling system extends the time of transition to a new temperature level, thereby increasing the change in the degree of conversion. The actual overheating of the reaction mixture with respect to the entering coolant temperature then is lower than the highest admissible value, whereby the reactor efficiency is unnecessarily lowered.

The aim of the present work was to seek how the reactor efficiency can be increased if a total model of the system is used.

## THEORETICAL

## MATHEMATICAL MODEL OF THE SYSTEM

The model is based on the concept of a perfectly stirred batch reactor and a perfectly stirred cooler. The dynamic behaviour of the system is described by three ordinary differential first order equations.

The mass balance of the reactor is described by the equation

$$dx/dt = r/c_{AO}, \quad (1)$$

the enthalpic balance of the reactor is described as

$$dT/dt = \Delta T_{ad}(dx/dt) - A_R(T - T_C), \quad (2)$$

where parameter  $A_R$  characterizes the intensity of heat exchange between the reaction mixture and cooler. Dissipation heat losses are disregarded. The enthalpic balance of the cooler can be described as

$$dT_C/dt = A_C(T - T_C) - C(T_C - T_{Ci}), \quad (3)$$

where parameter  $A_C$  is analogous to parameter  $A_R$  in Eq. (2) and constant  $C$  characterizes the mean residence time of coolant in the cooler.

For a case where the cooler has a low inertia, the mathematical model is simplified assuming that the cooler is in a pseudostationary state. Then the enthalpic balance of the reactor can be described by the equation

$$dT/dt = \Delta T_{ad}(dx/dt) - A_T(T - T_{Ci}), \quad (4)$$

where parameter  $A_T$  is

$$A_T = A_R C / (A_C + C). \quad (5)$$

## CONTROL ALGORITHM

The objective of nonisothermal control is keeping, by jump changes in the inlet temperature of coolant, the pseudostationary difference between the reaction mixture temperature and the entering coolant temperature in the maximum of the reaction mixture temperature at a preselected value given by the relation

$$T_{ps} - T_{Ci} = (T_{ps} - T_{Ci})_{\max} / S, \quad (6)$$

where  $S$  is a preselected safety coefficient value<sup>1</sup>.

The inlet temperature of coolant is set to a  $T_{Ci,n}$  level and stabilized there until the pseudostationary state is attained, *i.e.*, until the maximum temperature of the reaction mixture is recorded. In this moment the degree of conversion and the pseudostationary temperature difference  $T_{ps,n} - T_{Ci,n}$  are determined. The condition

$$dT/dt = 0 \quad (7)$$

is satisfied in the pseudostationary state, and therefore the relation

$$\Delta T_{ad}(r_i/A_T)(1-x)^k \exp [E(T_{ps} - T_{Ci})/(RT_{ps}T_{Ci})] = T_{ps} - T_{Ci} \quad (8)$$

holds true. The value of the adaptive parameter,  $r_i/A_T$ , is determined from this relation. The maximum temperature difference  $(T_{ps} - T_{Ci})_{max}$  must also meet the condition

$$\begin{aligned} (d/dT_{ps}) \{ \Delta T_{ad}(r_i/A_T)(1-x)^k \exp [E(T_{ps} - T_{Ci})/(RT_{ps}T_{Ci})] \} = \\ = (d/dT_{ps})(T_{ps} - T_{Ci}). \end{aligned} \quad (9)$$

For the determined degree of conversion, the highest temperature difference  $(T_{ps} - T_{Ci})_{max}$  is calculated from Eqs (8) and (9).

*Estimate of the new coolant temperature from the zeroth order model.* The new temperatures,  $T_{ps,n+1}$  and  $T_{Ci,n+1}$ , for a preselected safety coefficient, are obtained by solving the system of equations

$$(T_{ps} - T_{Ci})_{max}/[S(T_{ps,n} - T_{Ci,n})] = \exp [E(T_{ps,n+1} - T_{ps,n})/(RT_{ps,n}T_{ps,n+1})] \quad (10)$$

and

$$T_{Ci,n+1} = T_{ps,n+1} - (T_{ps} - T_{Ci})_{max}/S. \quad (11)$$

*Determination of the inlet coolant temperature from the total mathematical model.* The determined degree of conversion, measured reaction mixture temperature, measured coolant temperature in the cooler (in case that Eq. (3) is included in the model) and the inlet coolant temperature calculated from the zeroth order model serve as the starting values for the integration. The calculation is repeated with an inlet temperature increased or decreased by a preselected increment  $\Delta T_{Ci}$  according to whether the predicted course was safe or not. This is repeated until two inlet temperatures differing by  $\Delta T_{Ci}$  are found such that the course is safe at the lower temperature and hazardous at the higher. The highest safe inlet coolant temperature then is sought by the method of interval bisection until the difference between the two inlet temperatures tested is  $0.5^\circ\text{C}$ .

The predicted behaviour of the system is considered safe if a maximum appears on the simulated course of the reaction temperature, and hazardous if, during the integration, the calculated difference between the reaction temperature and the entering coolant temperature is higher than the  $(T_{ps} - T_{Ci})_{\max}/S$  value.

*Indication of the temperature maximum.* In the experimental application of the control algorithm, the reaction temperature data contain a noise component, which causes difficulties in the temperature maximum indication which is the starting point of the algorithm. An incorrect determination of the maximum can have unfavourable consequences.

For elimination of the effect of noise, a tolerance for the temperature maximum control  $\Delta T_{\text{reg}}$  is chosen such that it exceeds the noise bandwidth. The reaching of the maximum is indicated when the condition

$$T_{\text{obs}} < T_{\text{max}} - \Delta T_{\text{reg}} \quad (12)$$

is met; here  $T_{\text{max}}$  is the highest reaction mixture temperature recorded after changing the inlet coolant temperature and  $T_{\text{obs}}$  is the instantaneous reaction mixture temperature.

## EXPERIMENTAL

*Laboratory reactor.* A stirred batch reactor<sup>2,3,4</sup> with a reaction mixture volume of 560 cm<sup>3</sup> was used. The reaction heat was removed from the mixture by means of a cylindrical glass cooler with a volume of 7 cm<sup>3</sup> and exchange area of 39 cm<sup>2</sup>, submerged in the reaction mixture; the coolant flow rate was 6.4 cm<sup>3</sup> s<sup>-1</sup>. The inlet temperature of coolant was controlled with a thermostat whose time constant was about 30 s.

*Simulation apparatus.* The dependence of the reactor efficiency on the cooler inertia was studied on a simulation apparatus as described previously<sup>5,6</sup>. The apparatus comprised a batch reactor in which the exothermic chemical reaction was replaced by electric heating of a model system. The reactor was cooled by means of an external cooler whose inertia could be varied while keeping the heat exchange area, and practically also the heat-transfer coefficient, constant.

*Model reaction.* The model reaction was the highly exothermic oxidation of ethanol by hydrogen peroxide, homogeneously catalyzed by ferric ions<sup>3</sup>; its reaction enthalpy is  $-275 \text{ kJ mol}^{-1}$  with respect to H<sub>2</sub>O<sub>2</sub>. The starting concentrations of H<sub>2</sub>O<sub>2</sub> and C<sub>2</sub>H<sub>5</sub>OH were 2.04 and 0.84 mol . dm<sup>-3</sup>, respectively, the catalyst concentration was 1.77 mmol dm<sup>-3</sup>. The adiabatic temperature rise of the reaction was 130 K and activation energy 90 kJ mol<sup>-1</sup> with respect to H<sub>2</sub>O<sub>2</sub>. The kinetics of this reaction is described, with a sufficient accuracy, by the rate equation

$$dx/dt = 4 \cdot 10^{10} \exp(-10\,825/T)(1-x), \quad (13)$$

where  $x$  is the degree of conversion of hydrogen peroxide. This rate equation was used in the simulation apparatus control program. For studying the dependence of the increase in the reaction efficiency resulting from the use of the prediction method in question on the reaction order by using simulation calculations, the reaction order in this equation was varied while keeping the remaining parameters constant.

## RESULTS AND DISCUSSION

## EFFECT OF THE PREDICTION MODEL AND THE SAFETY COEFFICIENT ON THE HIGHEST OVERHEATING OF THE REACTION MIXTURE

The highest admissible pseudostationary overheating of the reaction mixture with respect to the temperature of entering coolant is determined by the condition

$$dT/dt = 0, \quad dT_c/dt = 0 \quad (14)$$

leading to the relation

$$(T - T_{Ci})_{\text{theor}} = (\Delta T_{\text{ad}}/A_T) (dx/dt), \quad (15)$$

where the left-hand term is the theoretical highest admissible temperature difference in the temperature maximum of the reaction mixture.

The actual temperature difference attained during the control (for a positive order reaction) is necessarily lower because surpassing this difference means control failure. The difference between the attained and the theoretical temperature differences is dependent upon the accuracy of the prediction model used, the safety coefficient, and the dynamic properties of the cooler. The effect of the two former factors was studied by simulation; the results are shown in Figs 1 and 2.

The maximum admissible overheating of the reaction mixture increases slightly with the degree of conversion. The actually attained overheating, for the zeroth

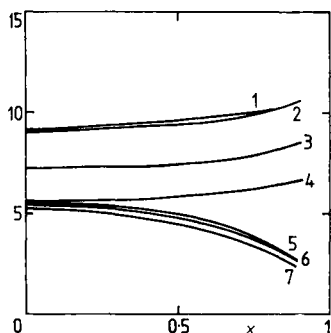


FIG. 1

Dependence of the maximum reaction mixture overheating on the degree of conversion for a 1st order reaction. 1 theoretical values, 2–4 total model, 5–7 zeroth order model.  $S = 1$  (2, 5), 1.25 (3, 6), 1.5 (4, 7)

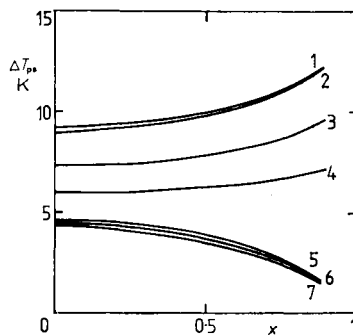


FIG. 2

Dependence of the maximum reaction mixture overheating on the degree of conversion for a 2nd order reaction. Curve labelling as in Fig. 1

order prediction model, decreases with the degree of conversion. This decrease is a consequence of the fact that the changes in the degree of conversion occurring on the transition from one inlet temperature of coolant to the next are disregarded; this is more significant at high degrees of conversion, when the reaction rate is very sensitive to changes in this quantity. A safety reserve thus forms when the zeroth order model is applied. The attained overheating is deeply below the admissible limit, and it depends only slightly on the chosen value of the safety coefficient.

When the total prediction model is used, the attained overheating of the reaction mixture increases with the degree of conversion and depends significantly on the chosen value of the safety coefficient. For the safety coefficient equal to unity the attained overheating is at the maximum admissible limit. The number of jump changes in the inlet coolant temperature necessary for reaching the given final degree of conversion decreases.

The magnitude of the maximum overheating of the reaction mixture has an effect on the reaction efficiency. Examples of comparison of the reactor efficiency for the exact model and the zeroth order prediction model are shown in Figs 3 and 4. The relative efficiency is expressed as the ratio of the reaction times necessary for reaching the final degree of conversion obtained by the zeroth order prediction model with  $S = 1$  ( $t_0$ ) and the exact model ( $t$ ). The use of the exact model thus enables the reactor efficiency to be increased. The increase is the higher the more the exact model departs from the zeroth order model; thus the difference increases with increasing reaction order and increasing final degree of conversion.

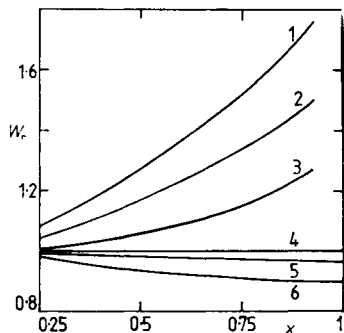


FIG. 3

Dependence of the relative reactor efficiency on the degree of conversion for a 1st order reaction. 1–3 total model, 4–6 zeroth order model;  $S = 1$  (1, 4), 1.25 (2, 5), 1.5 (3, 6)

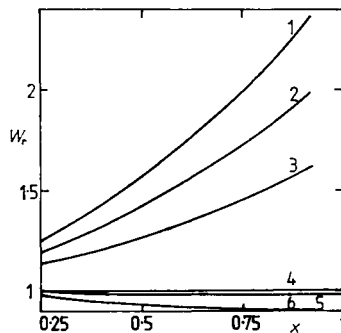


FIG. 4

Dependence of the relative reactor efficiency on the degree of conversion for a 2nd order reaction. Curve labelling as in Fig. 3

## EFFECT OF TEMPERATURE MEASUREMENT NOISE

The crucial problem of the control in the stable states is the temperature maximum indication. Error in this indication can result in control failure. If noise appears in the temperature measurement, the  $\Delta T_{\text{reg}}$  value, ensuring safety in the maximum indication, must be increased. The effect of the  $\Delta T_{\text{reg}}$  value on the reactor efficiency was examined experimentally using the laboratory reactor.

Figs 5 and 6 show the time dependences of the reaction temperature at safety coefficient values  $S = 1, 1.25,$  and  $1.67$  for  $\Delta T_{\text{reg}} = 0.1$  K (Fig. 5) and 1 K (Fig. 6). It is evident that while ensuring a safer indication of the reaction temperature maximum, a higher  $\Delta T_{\text{reg}}$  value slows down the indication and extends the time of transition from a temperature level to another, whereupon the reactor efficiency lowers. The prolongation of the temperature transition period is due to the fact that after the setting of the new inlet temperature of coolant the reaction temperature must first increase again by the  $\Delta T_{\text{reg}}$  difference and only then it begins to increase to above the value of the preceding maximum. During the reaction temperature decrease the reactor occurs in a pseudoisothermal regime, bringing about additional efficiency lowering.

Figs 7 and 8 show the course of the calculated and experimental maximum values of reaction mixture overheating. Fig. 9 is a plot of the dependences of the relative efficiency of the reactor on the degree of conversion for the three safety coefficient values. The measurement at  $\Delta T_{\text{reg}} = 0.1$  K served as the reference variant, and the values without subscripts are taken from measurements at  $\Delta T_{\text{reg}} = 1$  K. Both the attained overheating and the reactor efficiency are seen to decrease with increasing  $\Delta T_{\text{reg}}$ ; this decrease is highest at low degrees of conversion. In this stage of the reaction

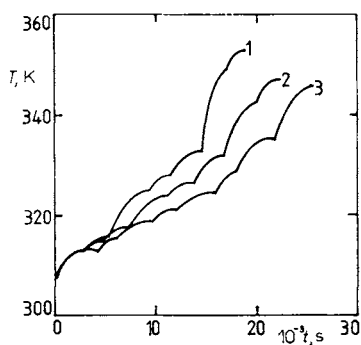


FIG. 5

Dependence of the reaction temperature on time for  $\Delta T_{\text{reg}} = 0.1$  K; total model,  $S$ : 1 1, 2 1.25, 3 1.67

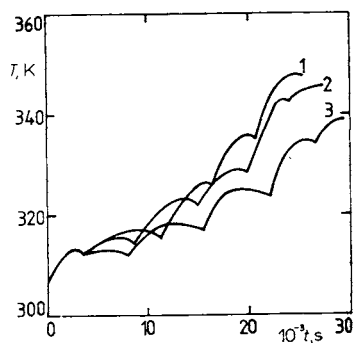


FIG. 6

Dependence of the reaction temperature on time for  $\Delta T_{\text{reg}} = 1$  K; total model,  $S$ : 1 1, 2 1.25, 3 1.67

the decrease in the reaction temperature by  $\Delta T_{\text{reg}}$  takes much longer times than at the end of the reaction; the reactor thus occurs in the unfavourable pseudoisothermal regime for a longer time.

The time dependences of the reaction temperature at the two  $\Delta T_{\text{reg}}$  values and  $S = 1$ , for the zeroth order prediction model and the total prediction model, are shown in Fig. 10. The reactor efficiency lowering brought about by increasing  $\Delta T_{\text{reg}}$

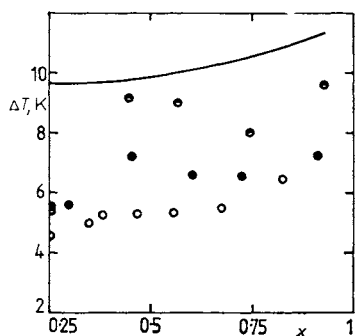


FIG. 7

Dependence of the maximum reaction mixture overheating on the degree of conversion at  $\Delta T_{\text{reg}} = 0.1$  K. — theoretical values; total model,  $S$ : ● 1, ● 1.25, ○ 1.67

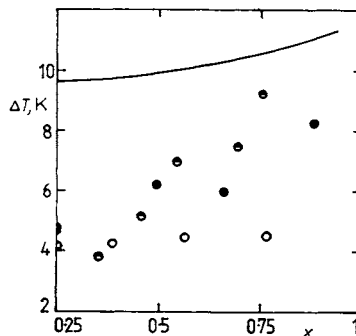


FIG. 8

Dependence of the maximum reaction mixture overheating on the degree of conversion at  $\Delta T_{\text{reg}} = 1$  K. Labelling as in Fig. 7

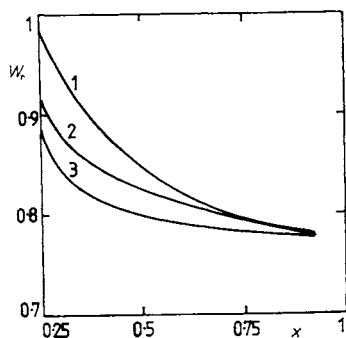


FIG. 9

Dependence of the reactor efficiency lowering resulting from the increase of  $\Delta T_{\text{reg}}$  from 0.1 K to 1 K on the degree of conversion; total model,  $S$ : 1 1, 2 1.25, 3 1.67

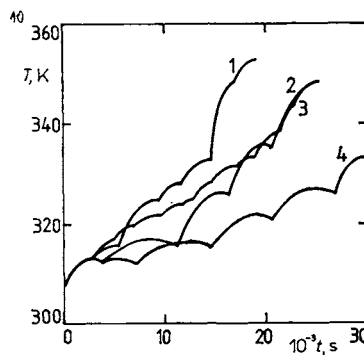


FIG. 10

Dependence of the reaction temperature on time at  $S = 1$  for the two prediction methods; 1, 2 total model, 3, 4 zeroth order model,  $\Delta T_{\text{reg}} = 0.1$  K (1, 3) and 1 K (2, 4)



from 0.1 K to 1 K is roughly made up for by the difference in the efficiency for the two prediction models.

#### EFFECT OF COOLER INERTIA

The effect of inertia of the cooler on the control was examined experimentally on the simulation equipment in which the inertia could be varied. The course of the reac-

TABLE I

Increase in the reactor efficiency resulting from the use of the mathematical model without and with the description of the cooler, with respect to the zeroth order model

$x$	Relative efficiency			
	mathematical model without cooler description, at $\tau_C/\tau_R$		mathematical model with cooler description, at $\tau_C/\tau_R$	
	0.03	0.13	0.03	0.13
0.50	1.30	1.29	1.30	1.30
0.75	1.62	1.60	1.64	1.63
0.90	1.85	1.80	1.89	1.89

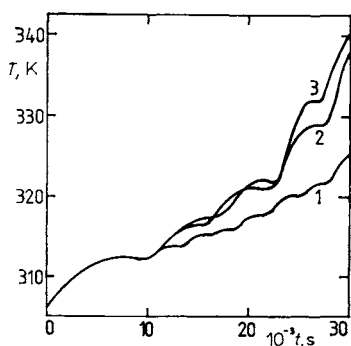


FIG. 11

Dependence of the reaction temperature on time at  $\tau_C/\tau_R = 0.03$ ; 1 zeroth order model, 2 total model without cooler description, 3 total model with cooler description

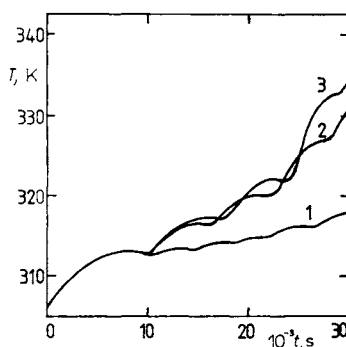


FIG. 12

Dependence of the reaction temperature on time at  $\tau_C/\tau_R = 0.13$ ; labelling as in Fig. 11

tion temperature was compared for the zeroth order prediction model and the total model both with and without the description of the dynamic behaviour of the cooler. The results, for the ratios of the cooler time constant to the reactor time constant  $\tau_C/\tau_R = 0.03$  and  $0.13$ , are shown in Figs 11 and 12, respectively. The safety coefficient was unity. Table I gives the relative reactor efficiency values for selected degrees of conversion; the measurements with prediction by means of the zeroth order model were taken as the reference ones.

The inclusion of the dynamic behaviour of the cooler in the mathematical model did not bring about any substantial improvement of efficiency. The efficiency increases slightly with increasing  $\tau_C/\tau_R$  ratio.

## CONCLUSIONS

The results indicate that owing to the increase in the accuracy of the prediction model, the reactor efficiency can be raised by using the total model. A relationship thus exists between the efficiency rise and the level of information about the process, entered into the algorithm. The zeroth order prediction model places less demands on the information about the process; the activation energy is sufficient datum, and the application of this model to positive order reactions also forms a safety reserve for the regime. The use of the total model makes it possible to attain a higher efficiency; higher demands, however, are put on the knowledge of the process and certainty of the prediction.

By the control method suggested the reactor efficiency can be increased without design alterations; only the reactor must be fitted with an equipment for varying the inlet coolant temperature.

## LIST OF SYMBOLS

$A$	parameter characterizing the the heat exchange intensity ( $s^{-1}$ )
$C$	parameter giving the inverse mean residence time of coolant residence in the cooler ( $s^{-1}$ )
$E$	activation energy ( $kJ\ mol^{-1}$ )
$k$	reaction order
$R$	gas constant ( $J\ mol^{-1}\ K^{-1}$ )
$r$	reaction rate ( $mol\ dm^{-3}\ s^{-1}$ )
$S$	safety coefficient
$T$	reaction mixture temperature (K)
$T_C$	coolant temperature in the cooler (K)
$T_{Ci}$	inlet temperature of coolant (K)
$\Delta T_{ad}$	adiabatic temperature rise of the reaction (K)
$\Delta T_{reg}$	tolerance of the temperature maximum control (K)
$t$	time (s)
$x$	degree of conversion of the principal component
$\tau$	time constant (s)

## Subscripts:

C	cooler
i	cooler inlet
max	maximum
n, n + 1	temperature level labelling
obs	observed value
ps	pseudostationary state
R	reactor/reaction mixture
theor	theoretical value
T	with respect to reactor if the cooler occurs in the pseudostationary state
0	reference conditions

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