TEMPERATURE CONTROL IN A CHEMICAL REACTOR THROUGH VARIABLE AREA OF THE HEAT TRANSFER SURFACE. EXPERIMENTAL VERIFICATION

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A possibility has been tested in the paper of the feed back control of temperature of the reaction mixture in a batch reactor with an exothermic reaction through the variable area of the cooling surface. The measurements were carried out in a laboratory reactor with a retractable cooler which was being immersed into the reaction mixture. The speed of motion of the cooler was sufficiently high permitting the process of immersion to be regarded as practically instantaneous. The aim of the control was to stabilize the set point temperature of the reaction mixture by a two-point controler. In dependence on the rate of response of the system to a change of the section variable either the ideal relay or the relay with hysteresis control algorithmus were used. The results of measurements showed that with the aid of a retractable cooler the temperature could be controlled safely even in those cases, in which the control by the variable flow rate of the coolant was unfeasible. The verification was carried out in the open-loop instable operating point of the reactor.

The usual action variables for temperature control in batch chemical reactors with an exothermic reaction are the flow rate and the inlet temperature of the coolant. A serious problem posed to this way of control is the inertia of the cooling system. The slow response of the cooling system is the result of the large thermal capacity of the cooling system. A possibility to circumvent this delay is through the use of the variable area of the heat exchange surface as the action variable. Simulation calculations on a mathematical model have shown[1] that this mode of control permits temperature control of the reactor even in its open-loop instable operating point.

This paper presents results of experimental verification of the temperature control in a batch reactor with an exothermic reaction by a retractable cooler immersed into the reaction mixture. The aim was to verify the results of the simulation under the practical conditions with various effects of system non-idealities such as the random noise in the temperature information and the time lag of the device sensing the temperature of the reaction mixture.

The aim of the control is to maintain the temperature of the reaction mixture in the reactor within a predetermined interval $T_s \pm T_{reg}$ by a controler with a two-level

output. At one level of the controller output the cooler in the reactor is being either immersed into the reaction mixture, or is permanently fully submerged. At the other level of the controler output, the cooler is being retracted from the mixture, or is permanently lifted outisde the reaction mixture.

The described method of control of the temperature of the reaction mixture has been compared with the conventional control by the variable flow rate of the coolant (the cooler being permanently immersed in the mixture).

The Control by the Retractable Cooler

The cooler was being immersed and retracted from the reaction mixture at a constant speed. The speed of motion of the cooler was so high that the process of immersion and retraction could be regarded as practically instantaneous. The response of the system to the change of the action variable was fast. The "relay with hysteresis" algorithm was applied for the control. Switching between both extreme positions was governed by the following conditions.

The cooler is being immersed into the mixture, or is permanently submerged if

$$T \geqq T_s + T_{reg}$$

or

$$dT/dt < 0 \quad \text{and} \quad T > T_s - T_{reg}.$$

The cooler is being retracted, or is permanently lifted outside the reaction mixture if

$$T \leqq T_s - T_{reg}$$

or

$$dT/dt < 0 \quad \text{and} \quad T < T_s - T_{reg}.$$

The band width of hysteresis of the controler (the band of insensitivity) in this case is defined by the value $h = T_{reg}$.

The Control by the Flow Rate of the Coolant

The cooler was permanently submerged into the reaction mixture. The temperature of the mixture was controlled by opening and closing the flow of the coolant into the cooler. For this mode of control the response of the system to the action variable was slow. The control was effected by the limit PD algorithm. This limit algorithm permits the highest safety margin of the operation to be accomplished. The switching of the flow rate of the coolant was governed by the following conditions.

The flow of the coolant is open if

$$dT/dt > 0 \quad \text{and} \quad T \geqq T_s - T_{reg}$$

The flow of the coolant is shut if

$$dT/dt \leqq 0 \quad \text{and} \quad T \leqq T_s + T_{reg} .$$

This algorithm is called the limiting or asymptotic PD algorithm for its derivative component has an infinite gain. The switching of the action variable is therefore effected instantaneously as soon as it has been safely established that the time derivative of the temperature, dT/dt, is non-zero. This means that the derivative has reached its threshold value distinguishable from the random noise. The derivative is taken to be positive if

$$T - T_{min} \geqq q$$

The derivative is taken to be negative, if

$$T_{max} - T \geqq q \quad \text{or for the time} \quad t_q \; |T - T_{max}| < q .$$

EXPERIMENTAL

As the model reaction we took a strongly exothermic oxidation of ethanol by hydrogen peroxide homogeneously catalyzed by Fe(III) ions[2]. The reaction may be described by a first-order reaction rate equation with respect to hydrogen peroxide

$$r = k_{st}C_{Ai}(1 - x) \; C_C \exp (-E/RT) .$$

The data pertaining to the reaction and the reaction conditions used in our experiments are summarized in Table I.

A scheme of the laboratory set-up is shown in Fig 1. The measurements were carried out in a mixed batch reactor (a Dewar vessel). The volume of the reaction mixture in the reactor was $5 . 10^{-4}$ m^3. A mixed cooler, which had a volume of $7 \cdot 7 . 10^{-5}$ m^3 and surface area of

TABLE I

Data pertaining to the model reaction

Parameter	k_{st}	E	ΔH	T_{ad}	c_{Ai}	c_{Bi}	c_C
Value	$7 \cdot 17 . 10^{13}$	90	$-275 \cdot 0$	$65 \cdot 0$	$1 \cdot 0$	$0 \cdot 516$	$0 \cdot 001 - 0 \cdot 002$

0.0133 m^2, was manufactured from a glass cylindrical vessel. The surface of the vessel was coated by a polystyrene foam whose variable thickness was utilized to adjust the value of the heat transfer coefficient between the reaction mixture and the coolant. The heat transfer coefficient ranged between 0.04 and $0.05 \text{ kW K}^{-1} \text{ m}^{-2}$. The cooler could be moved vertically by means of a two-point controlled servomotor. The speed of motion from one extreme position (fully submerged) into the other extreme position (fully retracted cooler) was constant and equal $3.6 \cdot 10^{-2} \text{ m s}^{-1}$. The time of travel of the cooler between both extreme positions was 6.5 seconds. The flow rate of the coolant (water) in the cooler could be varied between 0 and $5.41 \cdot 10^{-6} \text{ m}^3$. s^{-1}. The inlet temperature of the coolant was 286 K.

The temperature of the reaction mixture as well as the temperature of the coolant in the cooler were continuously monitored in the course of the experiment by thermocouples with time constants 3 and 5 seconds. Prior to the experiment the reaction mixture was heated to the reaction temperature and the reaction was started by feeding the solution of the catalyst In the case of the control by the flow rate of the coolant the initial temperature of the coolant in the cooler was equal to the initial temperature of the reaction mixture. In the case of the control by the mobile cooler the initial temperature of the coolant in the cooler was equal to the inlet temperature of the coolant. In both cases the control was initiated with switched-on cooling.
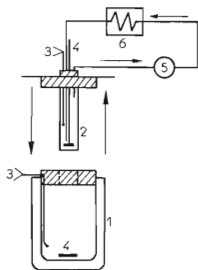

FIG. 1

Scheme of the laboratory batch reactor. 1 Reactor; 2 cooler; 3 thermocouples; 4 impeller; 5 circulation pump; 6 thermostat
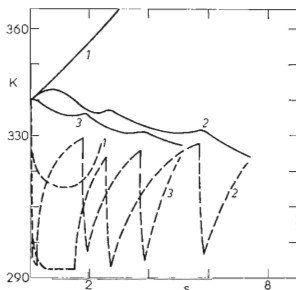

FIG. 2

Temperature control by the flow rate of coolant. The effect of the flow rate on the response of the system. $c_{\text{C}} = 0.001 \text{ kmol/m}^3$, $k_{\text{h}} = 0.05 \text{ kW/(Km}^2)$, $T_{\text{s}} = 340 \text{ K}$, $T_{\text{reg}} = 1 \text{ K}$, $q = 0.5 \text{ K}$, $t_{\text{q}} = 120 \text{ s}$; 1 $F_{\text{c}} = 3.2 \cdot 10^{-7} \text{ m}^3/\text{s}$; 2 $2.08 \cdot 10^{-6}$; 3 $5.41 \cdot 10^{-6}$. The solid line indicates the time course of the temperature of the reaction mixture; the broken line is the temperature of the coolant in the cooler

RESULTS AND DISCUSSION

The principal problem of a successful and chiefly safe control of the temperature of the reaction mixture in the batch reactor with an exothermic reaction is the removal of the reaction heat liberated in the course of the reaction. The difficulty of the control rests in that the requirements on the cooling rates vary with the degree of conversion of the reactants. Maximum cooling rates occur at the onset of the reaction. Due to the fact that the rate of the chemical reaction, and hence the rate of heat liberation depend exponentially on the temperature of the reaction mixture, the requirements on the cooling rate increase with the growing temperature at which the reactor is being operated. In those cases when the temperature of the reaction mixture is substantially higher than the temperature of the coolant in the cooler, it cannot be ruled out that the operating point (pseudo-stationary point) is open-loop instable. The temperature control then may significantly depend on the dynamic properties of the controled system, for instance, the inertia of the cooler. Elimination of this inertia poses a serious problem especially for a large capacity reactor[5]. An example may be batch polymeration reactors. Typical for these reactors is the relatively large volume of the cooling system (large thermal capacity) and unfavourable conditions for heat transfer due to the fouling of the exchange surfaces and extreme variations of viscosity of the reaction mixture in the progress of the reaction. The response of the cooling system to the change of the flow rate of the coolant or its inlet temperature is then slow. The results of previous studies[3-4] have shown that a two-level controller may be suitable for the temperature control of a batch reactor with an exothermic reaction.

If a continuously operating controller is used to control the temperature of the reaction mixture by the flow rate of the coolant, the response of the controlled system in the neighbourhood of the operating point (pseudo-stationary state) is approximately symmetric. This means that the response is identical for the decreasing and increasing direction of change of the flow rate of the coolant. The rate of response is determined by the properties of the system for the heat balance must hold

$$r_s(-\Delta H) V = F_{cs} c_{ps} \varrho_c (T_{cs} - T_{ci}) .$$

The flow rate of the coolant, which also determines the rate of the response of the cooling system to the change flow rate of the coolant is then a dependent variable and cannot be arbitrarily altered.

With a two-level controller which thoroughly shuts or opens the flow of the coolant the response is asymmetric. The response after closing the flow of the coolant is slow and its rate cannot be altered for it is given by the ratio of the thermal capacity of the reaction mixture and the cooling system. The response to the opening of the flow of the coolant may be fast an may be varied by the flow rate of the
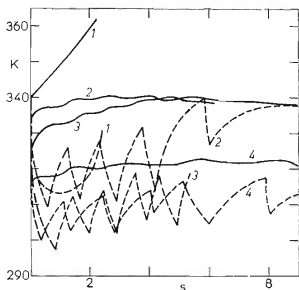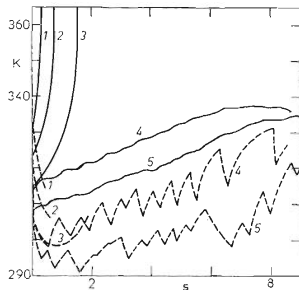
FIG. 3

Temperature control by the flow rate of coolant. The effect of the initial temperature of the reaction mixture on the response of the controlled system. $F_c = 3 \cdot 2 \cdot 10^{-7}$ m^3/s, $q = 1$ K, $t_q = 120$ s; 1 $T_s = 340$ K; 2 334; 3 326; 4 316. The remaining part of the caption is the same as for Fig. 2

FIG. 4

Temperature control by the flow rate of coolant. The effect of the initial temperature of the reaction mixture on the response of the controlled system. $c_C = 0 \cdot 002$ kmol/$/m^3$, $k_h = 0 \cdot 04$ kW/(km^2); 1 $T_s = 331$ K; 2 324; 3 315; 4 314; 5 $308 \cdot$. The remaining part of the caption is the same as for Fig. 3
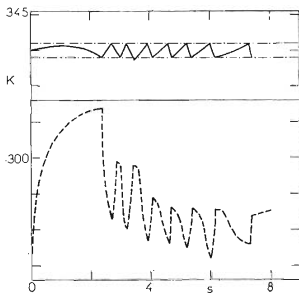
FIG. 5

Temperature control by the retractable coolder. $T_s = 340$ K, $F_c = 3 \cdot 2 \cdot 10^{-7}$ m^3/s, $M = 4 \cdot 2$. The remaining part of the caption is the same as for Fig. 2
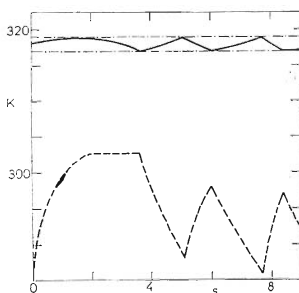
FIG. 6

Temperature control by the retractable cooler. $T_s = 318$ K, $F_c = 3 \cdot 2 \cdot 10^{-7}$ m^3/s, $M = 38 \cdot 0$. The remaning part of the caption is the same as for Fig. 4

coolant. The asymmetric response is favourable for the dangerous direction of temperature development is its growth. The two-level controller thus permits a high response of the system to be achieved in the hazardous direction. The slow response following the temperature drop is not so hazardous.

In practice the flow rate of the coolant, and hence the rate of the response, is limited by the maximum pumping capacity of the pump for the coolant, or the maximum permissible pressure drop in the cooling system.

The course of the response of the system during the control by the flow rate of the coolant is seen on Figs 2 − 4. Since the cooler had a large thermal capacity, the control was difficult and unreliable.

The large thermal capacity may be advantageous if the control is effected by a retractable cooler. The thermal capacity can be utilized as a "storage" of cold for checking the fast liberation of heat at the onset of the reaction. The dynamic properties of the control loop can be further selected independently on the properties of the controlled system and the chemical reaction by adjusting properly the speed of motion of the cooler. The speed of motion of the cooler was characterized in the previous simulation study[1] by the following dimensionless quantity

$$M = t_c/t_{reg}$$

i.e. by the ratio of the travel time of the cooler and the time required by the adiabatic course of the reaction to change the reaction temperature from $T_s − T_{reg}$ to $T_s + + T_{reg}$. If this dimensionless speed of the cooler exceeds unity, the immersion and retraction of the cooler may be regarded as instantaneous. The speed of motion of the cooler has then no effect on the magnitude of the control deviation of the temperature of the reaction mixture.

The experimental examples of temperature control of the reaction mixture under the conditions of high speed of motion of the cooler (M amounts to 4·2 to 38) are shown in Figs 5 and 6. While under identical reaction conditions and the flow rate of the coolant in the cooler, the control by the variable flow rate of the coolant was unsuccessful (Fig. 2 and 4), the control by the retractable cooler was reliable and of good quality. Since the response of the controlled system to the change of the action variable was swift we could do with a simple control algorithm "relay with hysteresis". If the width of hysteresis of the controller is smaller than the required accuracy of the temperature control (in the studied case both quantities had the same value), the use of the mentioned algorithm appears advantageous. It is so on the one hand for its relative simplicity, and, on the other hand, for its filtering of the random noise in the information about the temperature of the reaction mixture.

The results of experiments have proven the advantages of the retractable cooler for temperature control of the mixture in a batch reactor with an exothermic reaction. The advantages may be to a considerable extent offset by the increased costs expected

to be incurred by the more complicated construction of the cooling system of the reactor.

LIST OF SYMBOLS

c_{Ai}	initial concentration of hydrogen peroxide $(kmol/m^3)$
c_{Bi}	initial concentration of ethanol $(kmol/m^3)$
c_C	catalyst concentration $(kmol/m^3)$
c_{pa}	specific heat of the coolant $(kJ/kg\ K)$
E	activation energy (kJ/mol_A)
F_c, F_{cs}	flow rate of the coolant, flow rate corresponding to the pseudo-stationary state of the reactor (m/s)
h	width of hysteresis of the controller (K)
k_h	heat transfer coefficient $(kW/K\ m^2)$
k_{st}	constant in the reaction rate equation $(m^3/kmol_C\ s)$
M	dimensionless speed of motion of the cooler
q	present value of the temperature interval for the estimate of the sign of the temperature time derivative (K)
R	gas constant
r, r_s	reaction rate, reaction rate in the pseudo-stationary state of the reactor $(kmol_A/m^3\ s)$
T	temperature of the reaction mixture (K)
T_{ad}	adiabatic temperature rise (K)
T_c, T_{cs}	temperature of the coolant, temperature corresponding to the pseudo-stationary state of the reactor (K)
T_{ci}	inlet temperature of the coolant (K)
T_{min}, T_{max}	temperatures of the mixture on the beginning of the interval for the determination of the sign of dT/dt (K)
T_{reg}	present accuracy of control (K)
T_s	set point temperature, temperature of the pseudo-stationary temperature of the reaction mixture
t	reaction time (s)
t_c	travel time of the cooler (s)
$t_{reg} = T_{reg}c_{Ai}/(T_{ad}r_s)$	time of adiabatic course of the control process (s)
t_q	present value of the time interval for the estimate of the sign of dT/dt
V	volume of the reaction mixture (m^3)
x	degree of conversion of A
ΔH	reaction heat (kJ/mol_A)
ϱ_c	density of the coolant (kg/m^3)

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