CONTROL OF AN ADIABATIC CONTINUOUS REACTOR BY FEEDING CATALYST

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Received February 20th, 1979

Algorithms have been presented, analyzed and experimentally tested to stabilize the reaction temperature at constant inlet temperature and composition of the feed by controled dispensing of the catalyst. The information for the control element is the course of the reaction temperature. If the temperature of the reaction mixture is below the set point, the catalyst is being fed into the reactor at a constant rate. If the reaction temperature is higher the catalyst dispenser is blocked; dispensing of the catalyst is not resumed until the set point temperature has been reached again. The amount of catalyst added is a function of the duration of the switching cycle. The effect has been discussed of the form of this function on the course of the switching cycle. The results have been tested experimentally on a laboratory reactor controlled in an unstable steady state.

The aim of this work has been to work out and experimentally verify algorithms to maintain the reaction temperature by controlled dispensing of the catalyst through a device (action variable) which need not change continuously the feeding rate of the catalyst solution. At constant inlet temperature and composition of the reaction mixture the effected stabilization of the reaction temperature provides also for a stabilized outlet degree of conversion.

The paper is continuation of the previous communications[1,2] reviewing also the relevant literature.

THEORETICAL

Description of Algorithms and Their Designation

The aim of the control scheme is to maintain the reaction temperature in the neighbourhood of a set point T_s. The function of the regulator differs depending on whether the instantaneous temperature T is above or below the set point temperature.

The region $(T < T_s)$ or $(C_n < C_s)$: The reactor is fed with the catalyst solution at the rate F_{c1} until the reaction temperature has reached T_s.

Region $(T > T_s)$ or $(C_n > C_s)$: No catalyst solution is fed and hence its concentration in the reactor gradually decreases. As a consequence, the reaction temperature begins to decrease as well. At the instant when the reaction temperature hits T_s,

a volume of the catalyst solution, V_c, is added. The volume V_c is a function of the time Θ, during which the reaction temperature was in excess of T_s. Various types of functions $V_c(\Theta)$ tested for the control purpose are shown in Table I.

The Mathematical Model of the Reactor

The analysis was carried out for an exothermic reaction with the reaction rate being proportional to catalyst concentration. The reaction rate equation took the following form

$$r = k_0 c_{A0} c_c (1 - x) \exp \{(E \Delta T_{ad} x)/[R T_i (T_i + \Delta T_{ad} x)]\} . \qquad (1)$$

At constant inlet temperature and inlet composition the degree of conversion and catalyst concentrations during a switching cycle obeys the following equations

$$\frac{dx}{dt} = \frac{r}{c_{A0}} - \frac{F_i}{V_s} x \qquad (2)$$

$$\frac{dc_c}{dt} = - \frac{F_i}{V_s} (c_c - P c_s) \qquad (3)$$

with the initial conditions

$$t = 0 ; \quad x = x_s ; \quad c_c = c_n ,$$

$$t = t ; \quad x = x_s ; \quad c_c = c_e . \qquad (4)$$

TABLE I
Review of Studied Algorithms

Designation	$V_c(\Theta)$	Remark
I	$V_c = V_{c1}$	V_c independent of Θ
II	$V_c = F_{c2} \cdot \Theta$	V_c proportional to Θ
III	$V_c = V_{c1} + F_{c2} \cdot \Theta$	combination of I and II
IV	$V_c = \{V_s \cdot [C_{pn} - C_e(\Theta)]\}/(C_1 - C_{pn})$	initial catalyst concentration maintained at C_{pn}

Linearization of the Mathematical Model and Transformation to the Dimensionless Form

For the purpose of describing the behaviour of the reactor in the neighbourhood of the set point the properties of the system were expressed as linear functions of the deviations from steady state values. The effect of the degree of conversion and catalyst concentration on the progress of the reaction rate was expressed by the following relationship (Fig. 1)

$$r = r_s(1 + a\,\Delta x + \Delta c/c_s)\,. \tag{5}$$

After transformation to a dimensionless form the set of equations (2), (3) took the form

$$\frac{\mathrm{d}X}{\mathrm{d}\Theta} = (\pi - 1)\,X + (C - C_s) \tag{6}$$

$$\frac{\mathrm{d}C}{\mathrm{d}\Theta} = P - C \tag{7}$$

with the initial conditions

$$\Theta = 0\,; \quad X = X_n = 0\,; \quad C = C_n\,, \tag{8}$$

$$\Theta = \Theta\,; \quad X = 0\,; \qquad C = C_v\,.$$

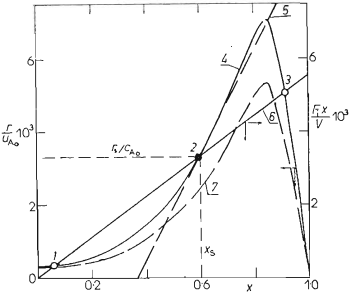

FIG. 1

Reaction Rate as a Function of the Degree of Conversion

1, *3* Stable steady states; *2* instable steady state; reaction rate as a function of the degree of conversion: *4* for $c_s = 1{\cdot}3 \,.\, 10^{-2}$ kmol/m^3 *5* linearized reaction rate; *6* operating line; *7* for $c_s = 10^{-2}$ kmol/m^3.

Integrating the set (6) and (7) a relation was obtained between the degree of conversion and catalyst concentration within a single switching cycle in the form

$$X = \frac{P - C}{\pi} + \frac{\lambda}{\pi} - \frac{1}{\pi}(P - C_n + \lambda - \pi X_n)\left[\frac{P - C}{P - C_n}\right]^{1-\pi}. \qquad (9)$$

The catalyst concentration at the end of the switching cycle, C_e (before volume V_c of catalyst was added), depends on the concentration of the catalyst at the onset of the switching cycle, C_n, by

$$0 = -C_e + P + \lambda - (P - C_n + \lambda)\left[\frac{P - C_e}{P - C_n}\right]^{1-\pi}. \qquad (10)$$

For the length of the switching cycle we may write

$$\Theta = \ln\left[\frac{C_n - P}{C_e - P}\right]. \qquad (11)$$

RESULTS AND DISCUSSION

The Determination of the Position and Stability of the Limit Cycle

The course of the switching cycle, n, is determined by the concentration at the onset of the cycle, C_n. Gradual changes of the initial catalyst concentration from cycle to cycle may therefore be determined graphically from the record of the dependence of the catalyst concentration at the end of the cycle, C_{n+1}, on the concentration at the beginning of the cycle, C_n. This dependence shall be designated $f(C_n)$. So long as it will be necessary to distinguish between the regions $T > T_s$ or $T < T_s$, we shall use the symbols $f_H(C_n)$ to refer to the former and the symbol $f_L(C_n)$ to the latter region. For the region $T > T_s$ it is advantageous to plot also the dependence of the catalyst concentration at the end of the cycle prior to the addition of the volume V_c; this function shall be designed as $C_e = f_e(C_n)$.

The limit cycles, approached gradually by the reactor regime, may be of two kinds:

1) The limit cycles formed by repetition of identical switching cycles. These are given by the intersect of the curve $f_H(C_n)$ with the diagonal given by the equation $C_{n+1} = C_n$.

These cycles are stable if we may write

$$\left|\frac{\mathrm{d}f_H(C_n)}{\mathrm{d}C_n}\right| < 1. \qquad (12)$$

2) The limit cycles with alternating switching cycles form the region $T > T_s$ and the region $T < T_s$. These limit cycles are given by the intersect of the function $f_H(C_n)$ and the function $f_L^{-1}(C_n)$, the latter being the inverse to the function $f_L(C_n)$.

These limit cycles are unstable if we may write

$$\frac{\mathrm{d}f_H(C_n)}{\mathrm{d}C_n} > 1 \quad \text{or} \quad \frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n} > 1 \quad \text{or} \tag{13a,b}$$

$$\left|\frac{\mathrm{d}f_H(C_n)}{\mathrm{d}C_n}\,\frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n}\right| > 1 . \tag{13c}$$

In case that the left hand side of the inequality (*13c*) equals unity the limit cycle is stable if, and only if, we may write in its neighbourhood

$$f_H(C_n) > f_L^{-1}(C_n) . \tag{13d}$$

The Switching Cycle in the Region $T < T_s$

The course of the switching cycle depends on whether the set point is a stable or an unstable steady state of the reactor. The set point is stable provided

$$\frac{\mathrm{d}r}{\mathrm{d}x} < Fc_{A0}/V .$$

From the linearized model of the reactor it follows that the function $f_L(C_n)$ possesses the following properties:

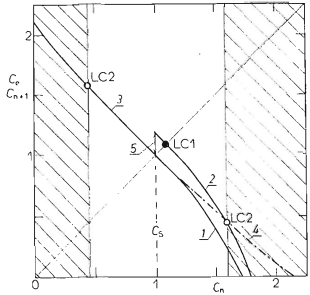

FIG. 2

Algorithm I — Course of Functions $f(C_n)$ and $f_e(C_n)$

1 Function $f_e(C_n)$; *2* function $f_H(C_n)$; *3* function $f_L(C_n)$ for $P = 4$; *4* function $f_L^{-1}(C_n)$; *5* constant addition of catalyst V_{c1}; /// region of instability; LC1, LC2 limit cycles.

For the stable set point

$$0 > \frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n} > -1\,; \quad \lim_{C_n \to 1} \frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n} = -1\,; \quad \frac{\mathrm{d}^2 f_L(C_n)}{\mathrm{d}C_n^2} < 0 \qquad (14a)$$

For an unstable set point

$$\frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n} < -1\,; \quad \lim_{C_n \to 1} \frac{\mathrm{d}f_L(C_n)}{\mathrm{d}C_n} = -1\,; \quad \frac{\mathrm{d}^2 f_L(C_n)}{\mathrm{d}C_n^2} > 0\,. \qquad (14b)$$

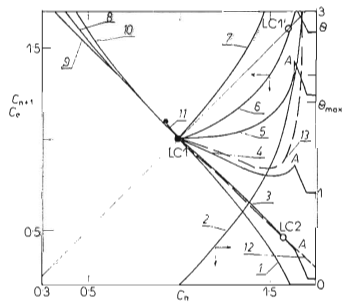

FIG. 3

Algorithm II — Effect of Regulator Parameters F_{c1} and F_{c2} on the Course of Functions $f_L(C_n)$ and $f_H(C_n)$

1 Function $f_e(C_n)$; *2* course of function $\Theta(C_n)$; *3*—*7* course of function $f_H(C_n)$ for various parameters F_{c2}: *3* $F_{c2} = 0{\cdot}57 \,.\, 10^{-6}$ m^3, *4* $1{\cdot}66 \,.\, 10^{-6}$ m^3, *5* $3{\cdot}03 \,.\, 10^{-6}$ m^3, *6* $4{\cdot}0 \,.\, 10^{-6}$ m^3, *7* $5{\cdot}9 \,.\, 10^{-6}$ m^3; *8*—*11* courses of function $f_L(C_n)$ for various values of parameter *P*: *8* $P = 3$, *9* $P = 8$, *10* $P = 2$, *11* $P = 1{\cdot}1$; *12* function $f_L^{-1}(C_n)$ for $P = 8$; point *A* marks position of maximum addition of the catalyst, V_{max}; LC1, LC1′, LC2 limit cycles.

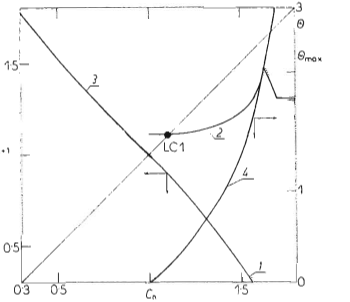

FIG. 4

Algorithm III — Courses of Function $f(C_n)$ and $f_e(C_n)$

1 Function $f_e(C_n)$; *2* function $f_H(C_n)$ for $F_{c2} = 3{\cdot}03 \,.\, 10^{-6}$ m^3; *3* function $f_L(C_n)$ for $P = 3$; *4* function $\Theta(C_n)$.

Examples of the function $f_{IL}(C_n)$ are shown in Figs 3 and 6. In the course of the switching cycle the catalyst concentration is increased above the value corresponding to the set point. The results show that this undesirable overdosing diminishes with increasing feed rate of the catalyst F_{c1}. In a system without time delays in the control loop it is therefore advantageous to use a maximum feed rate F_{c1}. In real systems under the effect of time delay, a compromise must be sought.

The Switching Cycle in the Region $T > T_s$

From the linearized model there follows that the dependence of the catalyst concentration prior to the addition of the volume V_c has the following properties:

For the stable set point:

$$0 > \frac{\mathrm{d}f_e(C_n)}{\mathrm{d}C_n} > -1\,; \quad \lim_{C_n \to 1} \frac{\mathrm{d}f_e(C_n)}{\mathrm{d}C_n} = -1\,; \quad \frac{\mathrm{d}^2 f_e(C_n)}{\mathrm{d}C_n^2} > 0\,. \qquad (15a)$$

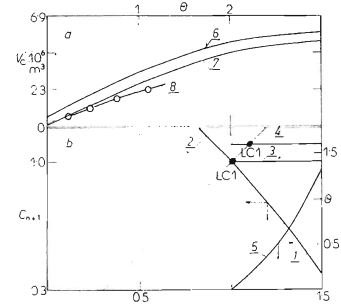

FIG. 5

Algorithm IV

a) Volume of catalyst added as a function of the length of the switching cycle. *b*) course of functions $f(C_n)$, $f_e(C_n)$ and $\Theta(C_n)$ *1* function $f_e(C_n)$; *2* $f_L(C_n)$ for $P = 5$; *3* $f_H(C_n)$ for $C_{pn} = C_s$; *4* $f_H(C_n)$ for $C_{pn} > C_s$; *5* function $\Theta(C_n)$; *6* V_c as a function of Θ for the straight line *4*; *7* V_c as a function of Θ for the straight line *3*; *8* V_c as a function of Θ obtained experimentally.

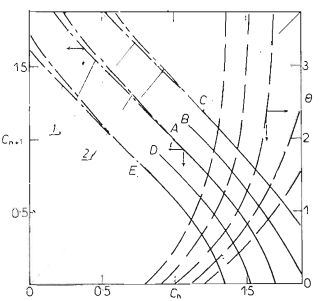

FIG. 6

The Effect of Parameter Lambda on the Course of Functions $f_e(C_n)$, $f_L(C_n)$ and $\Theta(C_n)$

Curves originating at *A*: $\lambda = 1{\cdot}693$, $\pi = 2{\cdot}4423$; curves originating from *B*, *C*, *D*, *E* — change of parameter lambda by +10%, +20%, —10%, —20%; ——— function $f_e(C_n)$; - - - - - function $\Theta(C_n)$; - · - · - · - function $f_L(C_n)$ for: *1* $P = 8$, *2* $P = 3$.

For an unstable set point:

$$\frac{\mathrm{d}f_e(C_n)}{\mathrm{d}C_n} < -1\,; \quad \lim_{C_n \to 1} \frac{\mathrm{d}f_e(C_n)}{\mathrm{d}C_n} = -1\,; \quad \frac{\mathrm{d}^2 f_e(C_n)}{\mathrm{d}C_n^2} < 0\,. \tag{15b}$$

The function $f_H(C_n)$ equals the sum of the functions $f_e(C_n)$ and $V_c(C_n)$.

Algorithm I

For the added volume of the catalyst solution we write: $V_c = V_{c1}$; where V_{c1} is a constant. In region $C_n > C_s$ we thus have

$$f_H(C_n) = C_c + V_{c1} C_1 / V_s\,. \tag{16}$$

An example of the determination of the limit cycle for the control in an unstable operating point is shown in Fig. 2. It is apparent that there exists only one steady

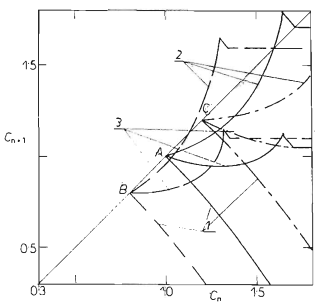

FIG. 7

Algorithm II — Effect of Parameter Lambda on the Course of Function $f_H(C_n)$

Curves originating from *A*: $\lambda = 1{\cdot}693$, $\pi = 2{\cdot}4423$; curves originating from points *B*, *C* change of parameter lambda by —20%, +20% *1* $f_e(C_n)$; *2* $f_H(C_n)$ for $F_{c2} = 4{\cdot}0 \,.\, 10^{-6}$ m^3; *3* $f_H(C_n)$ for $F_{c2} = 2{\cdot}4 \,.\, 10^{-6}$ m^3.

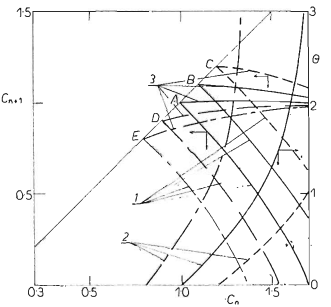

FIG. 8

Algorithm IV — Effect of Parameter Lambda on the Course of Functions $f_H(C_n)$ and $\Theta(C_n)$

Curves originating from *A*: $\lambda = 1{\cdot}693$, $\pi = 2{\cdot}4423$; curves originating from points *B*, *C*, *D*, *E* change of lambda by +10%, +20%, —10%, —20%; *1* $f_e(C_n)$; *2* $\Theta(C_n)$; *3* $f_H(C_n)$.

state consisting of only one switching cycle (LC1) given by the intersect of the curve $f_H(C_n)$ with the diagonal ($C_{n+1} = C_n$). From the equations (*15*), (*12*) and (*16*) there follows that the state is stable if, and only if, the control is that in a stable operating point. In case of an unstable operating point the limit cycle is unstable. Apart from this there exist still a limit cycle LC2 with alternating cycles one from the region $C_n > C_s$ and one from $C_n < C_s$. The limit cycle LC2 is given by the intersects of the curves $f_H(C_n)$ and $f_L^{-1}(C_n)$. From the mathematical model there follows that for an unstable state of the reactor the criterial condition (*13*) is always fulfilled; the limit cycle LC2 is therefore also unstable and represents the limit of the domain of attraction. Once the reactor has reached this region it remains there and forms a limit cycle of a complex aperiodic structure.

From the practical point of view the algorithm appears advantageous for its simplicity although it does exhibit some unfavourable properties. The smaller the value V_{c1}, the closer the limit cycle to T_s while simultaneously the domain of attrac-

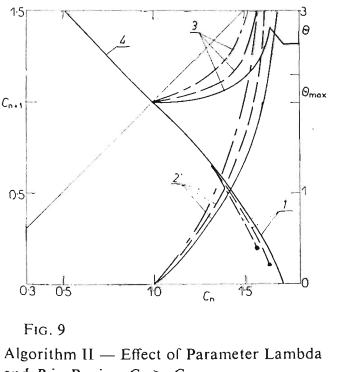

FIG. 9

Algorithm II — Effect of Parameter Lambda and P in Region $C_n > C_s$

——— $\lambda = 1{\cdot}693$; $P = 0$; $\pi = 2{\cdot}4423$; - - - - $\lambda = \lambda$ —10%; $P = 0{\cdot}1$; -·-·-·- $\lambda = \lambda$ — 20%; $P = 0{\cdot}2$; *1* $f_c(C_n)$, ● transition of the reactor into the cold state; *2* $\Theta(C_n)$; *3* $f_H(C_n)$ for $F_{c2} = 3{\cdot}03 \cdot 10^{-6}$ m^3; *4* $f_L(C_n)$ for $P = 5$.

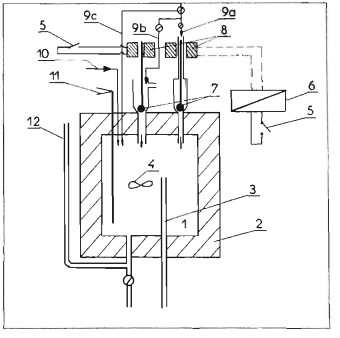

FIG. 10

Scheme of Apparatus

1 Reactor; *2* polystyrene foam insulation; *3* overflow; *4* stirrer; *5* switch; *2* time relay; *7* ball valve; *8* coil; *9* catalyst dispenser: *a*) constant volume V_{c1}, *b*) V_c proportional to Θ, *c*) feed at constant rate F_{c1}; *10* feed of reaction mixture; *11* thermocouple; *12* gauge.

tion grows narrower. The requirement for a wide domain of attraction leads to an increased V_{c1}, which causes the limit cycle to depart from T_s with impaired quality of regulation.

An example of experimental test of the algorithm is presented in Fig. 11*c*. The measurements were carried out under the conditions given in Experimental.

Algorithm II

For the added volume of the catalyst solution we write

$$V_c = F_{c2}\Theta \,. \tag{17a}$$

The results of measurement indicate that the regulator following the algorithm II in Eq. (*17a*) exhibits excessively narrow domain of attraction (an example of the function $f_H(C_n)$, defined in terms of Eq. (*17a*), is shown in Fig. 3 – curve *13*). The domain of attraction may be widened by diminishing the volume of the catalyst added:

$$V_c = F_{c2}\Theta \quad \text{for} \quad \Theta < \Theta_{max} \,, \tag{17b}$$

$$V_c = V_{max} = F_{c2}\Theta_{max} \quad \text{for} \quad \Theta \geqq \Theta_{max} \,.$$

The value of the constant V_{max} or Θ_{max} influences position of the point A as well as the unstable limit cycle LC1′ (Fig. 3) and thereby considerably expands the domain of attraction.

The effect of the constant F_{c2} *on stability of the limit cycle.* For this mode of control there exists the limit cycle LC1 directly in the operating point. The function $f(C_n)$ has in this point a discontinuous derivative. From the criterion of stability, Eq. (*13*), and the mathematical model there followed that for the control in an unstable operating point for $F_{c2} = 0$, the state LC1 is unstable, but for an arbitrarily small F_{c2} changes to the stable state. In addition, an unstable limit cycle LC2 forms, which restricts the domain of attraction. The state LC2 is determined by the intersect of the functions $f_H(C_n)$ and $f_L^{-1}(C_n)$ (Fig. 3). The domain of attraction initially widens with increasing value of F_{c2} (Fig. 3, curves *3*–*5*), the unstable cycle LC2 vanishes. Later on the domain of attraction is being confined by the unstable limit cycle LC1′, given by the intersect of the function $f_H(C_n)$ with the diagonal (Fig. 3, curve *6*). This unstable state LC1′ approaches with increasing value of F_{c2} the operating point LC1 until they completely coincide which makes the operating point instable (Fig 3, curve *7*).

If the state LC1 is stable, the ideal regulator tends to the state with alternating infinitely small additions of the catalyst solution at the rates F_{c1} and F_{c2} so as to

make the concentration of the catalyst within the reactor equal C_s. For a real regulator a limit cycle appears, given by the dynamic properties of the control loop. It is therefore advantageous to adjust the constant F_{c2} and Θ_{max} so as to have in the largest possible neighbourhood of the operating point the value $f_H(C_n)$ close to C_s. This ensures rapid convergence to the operating point and simultaneously provides for low sensitivity to the changes of system parameters.

Examples of experimental tests of the algorithm II for various values of parameters of the regulator are given in Fig. 12. Since the laboratory reactor was under the effect of various time-delay terms, a somewhat smaller values of the parameter $F_{c1} = 3{\cdot}3 \cdot 10^{-8}$ m^3/s had to be used in order to achieve a successful regulation.

Algorithm III

This algorithm is a combination of the algorithms I and II. The amount of the catalyst added is defined by the following equations

$$V_c = V_{c1} + F_{c2}\Theta \quad \text{for} \quad \Theta < \Theta_{max}\,,$$
$$V_c = V_{c1} + V_{max} = V_{c1} + F_{c2}\Theta_{max} \quad \text{for} \quad \Theta \geqq \Theta_{max}\,. \tag{18}$$

This algorithm provides for the existence of a stable limit cycle LC1 for $C_n \neq C_s$. In the steady state the regulator operates only in region $C_n > C_s$. An example of the $f(C_n)$ is given in Fig. 4.

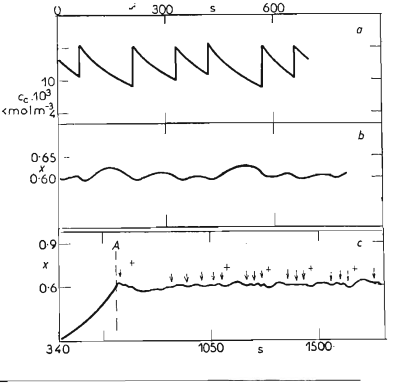

FIG. 11

Algorithm IV — Experimental Verification
a Dependence of catalyst concentration on time; *b* corresponding time course of the degree of conversion; $C_{pn} = 1{\cdot}2$; dependence $V_c(\Theta)$ shown in Fig. 5 — curve *8*; *c* algorithm I experimental verification; at point *A* start of feeding reaction mixture; ↓ addition of volume V_{c1} of catalyst; + catalyst fed at the rate $F_{c1} = 0{\cdot}06 \cdot 10^{-6}$ m^3/s; $c_1 = 0{\cdot}5$ kmol/m^3; $V_{c1} = 1{\cdot}2 \cdot 10^{-6}$ m^3.

Algorithm IV

For this mode of control the relationship between the length of the switching cycle and the amount of the catalyst added, V_c, was taken so as just balance the decrease of the concentration during the switching cycle

$$V_c = V_s[C_{pn} - C_e(\Theta)]/(C_1 - C_{pn}) . \qquad (19)$$

The relationship (*19*) must be further specified with the aid of the mathematical model for the concentration C_e depends on the length of the switching cycle, Θ, and the initial concentration C_{pn}, according to Eq. (*11*). An example of this dependence is given in Fig. 5*a* – curves *6*, *7*.

This mode of control ensures rapid convergence in the whole range of concentrations C_n. Depending on the chosen value of C_{pn} there may exist a stable limit cycle LC1 either directly in the operating point ($C_{pn} = C_s$), or in region $C_n > C_s$ ($C_{pn} > C_s$) (Fig. 5*b*, curve *3*, *4*). An example of experimental test for the case $C_{pn} > C_s$ is presented in Fig. 11*a*,*b*.

The Effect of Parameter Changes on Stability

The inlet disturbances to which the reactor is exposed may be of different origin: The feed rate may fluctuate, the content of catalytic poisons as well as the content of active substance in the feed may vary, or eventually, the constants of the regulator may also drift (F_{c1}, F_{c2}, V_{c1}, V_{max}) (it is assumed that the inlet temperature and con-

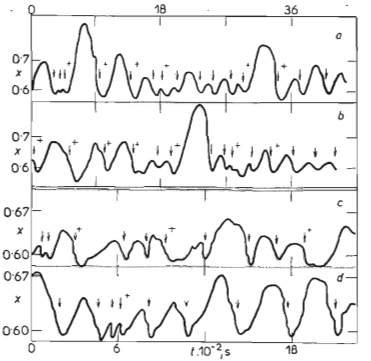

FIG. 12

Algorithm II. Experimental Tests for Various Values of Regulator Parameters

a, *b*—$F_{c1} = 0{\cdot}075 \cdot 10^{-6}$ m^3/s; $V_{max} = 4{\cdot}5 \cdot 10^{-6}$ m^3; $F_{c2} = a)$ $2{\cdot}69 \cdot 10^{-6}$ m^3, *b*) $3{\cdot}58 \cdot 10^{-6}$ m^3; *c*, *d*— $F_{c2} = 3{\cdot}58 \cdot 10^{-6}$ m^3; $V_{max} = 4{\cdot}8 \cdot 10^{-6}$ m^3; $F_{c1} = c)$ $0{\cdot}033 \cdot 10^{-6}$ m^3/s, *d*) $0{\cdot}068 \cdot 10^{-6}$ m^3/s; ↓ addition of volume V_c of catalyst; + feeding of catalyst at the rate F_{c1}.

centration are constant). By a suitable transformation of the mathematical model into the dimensionless form the effect of all these disturbances may be lumped into a single dimensionless parameter λ. The value of this parameter grows if the feed rate, or the content of catalytic poisons in the feed rate, or the content of catalytic poisons in the feed increase, if the concentration of the fed catalyst solution decreases, or eventually, if the values of the parameters of the regulator decrease.

In order to find the range of changes in which individual algorithms ensure stability of the regulation, the effect was assessed of the parameter lambda on the course of the functions $f_H(C_n)$, $f_L(C_n)$, $f_e(C_n)$ and $\Theta(C_n)$.

It was established that increased values of the parameter lambda diminish the duration of the swithing cycle Θ, shift the operating point C_s toward the hinger values, decrease the slope $df_h(C_n)/dC_n$, and, in case that the stable limit cycle LC1 does not coincide with the operating point C_s, shift this limit cycle toward the operating point. The situation is reverse following a decrease of the parameter lambda. Examples obtained by simulation are shown in Figs 6–9.

The domain of attraction for $C_n > C_s$ is considerably sensitive to the change of the parameters while for $C_n < C_s$ the parametric sensitivity is considerably weaker. It is apparent that from the standpoint of the parametric sensitivity the regulation functions may be chosen so as to maintain stability of the system even after a change of the parameter lambda by $\pm 20\%$. In the selection of the regulation function $f_H(C_n)$ and the adjustement of the regulator parameters one has to regard as the least favourable alternative the minimum possible value of the parameter lambda. With the changes induced by the growth of this parameter copes the regulation function $f_L(C_n)$ in region $C_n < C_s$.

The mathematical model was solved numerically on the TESLA 200 computer; the values of parameters were determined from the constants obtained from kinetic measurements (Table II).

EXPERIMENTAL

As the model reaction we used oxidation of ethanol by hydrogen peroxide homogeneously catalyzed by ferric ions in a laboratory adiabatic continuous stirred tank reactor described in ref.[2] (Fig. 10).

The regulator realizing the algorithm I consisted of a relay opening for a fixed period of time the feed of the catalyst into the reactor. The functions of the regulators simulating the algorithms II and III were realized by a storage tank of adjustable volume V_{max} put upstream into series with the reactor. The storage tank was filled during the switching cycle at the rate F_{c2} by the catalyst solution of concentration C_1. At the time when the temperature T_s was reached the content of the tank was discharged into the reactor.

Measuring method. The reactor was brought into the steady state in the batchwise mode of operation. After filling the reactor with the reaction mixture (composition corresponding to the feed, catalyst concentration $c_c = 1{\cdot}6 \cdot 10^{-2}$ kmol/m^3) to the operating volume, the measurement of the temperature of the reaction mixture was started. Having reached a temperature close to, but

higher than the steady state temperature, the feeding of the reactor was actuated and the algorithm I was used to bring the reactor into the steady state. Only then did the proper control of the reactor begin following one of the selected algorithms I—IV (examples are shown in Figs 11, 12).

The experimental control of the reactor was realized under the condition of three possible steady states in the reactor (Fig. 1). The values of the inlet stream parameters were in all experiments constant and are summarized in Table II. The state corresponding to these conditions is an unstable steady state characterized by the degree of conversion $x_s = 0{\cdot}6$, by the reaction volume of the mixture $V_s = 2{\cdot}15 \cdot 10^{-4}$ m^3 and catalyst concentration $c_s = 1{\cdot}3 \cdot 10^{-2}$ kmol/m^3.

CONCLUSION

The results of this work proved that the proposed algorithms may be used for the control of the reaction temperature even in an unstable operating point. The control, however, may lead to the limit cycle when the temperature oscillates in close neighbourhood of the operating point. In real systems the magnitude of the temperature

TABLE II

Applied Values of Constants and Parameters

Constant	Value	Dimension
	Parameters at the reactor inlet	
T_i	293	K
F_i	$1{\cdot}2 \cdot 10^{-6}$	m^3 s^{-1}
c_{A0}	1·2	kmol m^{-3}
c_{B0}	0·62	kmol m^{-3}
k_0	$6{\cdot}42 \cdot 10^{-3}$	m^3 kmol$_C^{-1}$ s^{-1}
	Parameters of instable steady state	
x_s	0·6	—
T_s	333	K
V_s	$2{\cdot}15 \cdot 10^{-4}$	m^3
c_s	$1{\cdot}3 \cdot 10^{-2}$	kmol m^{-3}
r_s/c_{A0}	$3{\cdot}3 \cdot 10^{-3}$	s^{-1}
	Other used constants	
c_1	0·5	kmol m^{-3}
E	90	kJ mol$_A^{-1}$
ΔT_{ad}	67·5	K
R	8·314	J mol^{-1} K^{-1}

from the set point depends on the time delay and the level of noise in the control loop. For a successful control the effect of these nonidealities must be checked.

From the results there follows that for the control in an unstable point a successful control may be ensured only if the inlet parameters (rate of catalyst feed, concentration and activity of the catalyst solution, feed rate of the reaction mixture, operating volume of the reactor) vary only within $\pm 30\%$ rel. In case of greater fluctuations a regulator with adaptive feature would have to be used.

LIST OF SYMBOLS

$a = \Delta T_{ad} \cdot E/(R \cdot T_s^2) - 1/(1 - x_s)$ dimensionless constant in equation 5
$C = c_c/c_s$ dimensionless catalyst concentration
C_1 dimensionless concentration of dispensed catalyst solution
C_e dimensionless catalyst concentration at the end of switching cycle
C_{pn} dimensionless constant catalyst concentration at the beginning of the switching cycle for algorithm IV
c_{A0} inlet concentration of the key component H_2O_2 (kmol/m^3)
c_{B0} inlet concentration of ethanol (kmol/m^3)
c_c catalyst concentration (kmol/m^3)
c_s steady state catalyst concentration (kmol/m^3)
$\Delta c = c_c - c_s$ deviation of catalyst concentration from steady state value (kmol/m^3)
E activation energy (J mol$_A^{-1}$)
F_{c1} feed rate of catalyst in region $T < T_s$ (m^3 s^{-1})
F_{c2} parameter of regulator II, III (m^3)
F_i feed rate of reaction mixture (m^3 s^{-1})
$f(C_n)$ functional valuee of $C_{n+1} = f(C_n)$
$f_e(C_n)$ functional values of $C_e = f(C_n)$ in region $T > T_s$
$f_H(C_n)$ functional values of $C_{n+1} = f(C_n)$ in region $T > T_s$
$f_L(C_n)$ functional values of $C_{n+1} = f(C_n)$ in region $T < T_s$
$f_L^{-1}(C_n)$ function inverse to $f_L(C_n)$
k_0 reaction rate constant at temperature T_i (m^3 kmol$_C^{-1}$ s^{-1})
LC1 limit cycle consisting of single switching cycle
LC2 limit cycle consisting of two switching cycles
$n, n+1$ index of the switching cycles
$P = F_{c1}c_1/(F_i c_s)$ constant of the regulator for region $T < T_s$
R gas constant (J mol^{-1} K^{-1})
r reaction rate (kmol$_A$ m^{-3} s^{-1})
T temperature of the reaction mixture (K)
ΔT_{ad} adiabatic temperature rise (K)
t time (s)
V_c dispensed volume of the catalyst solution (m^3)
V_{c1} constant dispensed volume of catalyst solution for algorithms I, III (m^3)
V_s operating volume of reactor (m^3)
$X = (x - x_s)/x_s$ dimensionless conversion of the key component
x degree of conversion of the key component
$\Delta x = x - x_s$ deviation of the degree of conversion from steady state value
$\lambda = (C_s - P)\,\pi/(\pi - 1)$ dimensionless parameter of mathematical model

$\pi = x_s \,.\, a$ dimensionless parameter of mathematical model
$\Theta = t \,.\, F_i / V_s$ dimensionless time-length of switching cycle

Subscript

A key component H_2O_2
B ethanol
C catalyst
i inlet
max maximum value
min minimum value
$n, n + 1$ at the beginning of the n-th, or $(n + 1)$-th switching cycle
s operating (set) point

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Translated by V. Staněk.