

## A SIMULATION APPARATUS FOR THE EXPERIMENTAL STUDY OF BATCH REACTOR CONTROL METHODS

Josef HORÁK and Petr BERÁNEK

*Department of Organic Technology,  
Prague Institute of Chemical Technology, 166 28 Prague 6*

Received February 7th, 1985

---

A simulation apparatus for the experimental study of the methods of control of batch reactors is devised. In this apparatus, the production of heat by an exothermic reaction is replaced by electric heating controlled by a computer in a closed loop; the reactor is cooled with an external cooler whose dynamic properties can be varied while keeping the heat exchange area constant. The effect of the cooler geometry on its dynamic properties is investigated and the effect of the cooler inertia on the stability and safety of the on-off temperature control in the unstable pseudostationary state is examined.

---

A number of technical problems are encountered in the experimental study of the control of chemical reactors and in the associated research and education activities. Quite a serious problem for large-size reactors, for instance, is the cost of chemicals and product disposal. Also, safety precautions must be made to avoid harmful control failure. Moreover, in actual chemical reactors the study is constrained by the fact that the properties of the chemical reaction involved, such as its activation energy or reaction order, cannot be varied. These problems can be circumvented by using a device in which the heat evolution from a chemical reaction is imitated by electric heating of an inert liquid (*e.g.*, water) inside the reactor, the heating being controlled by a computer according to the entered rate equation of the reaction. The heat from the inert reactor filling is removed by a real cooler. This approach combines the advantages of a total simulation with the practical aspects of the experimental work.

An apparatus of this kind is described in the present paper. A similar equipment has been devised by Mukesh and Cooper<sup>1</sup>. The aim of our work was to design an apparatus where the dynamic properties of the cooler can be varied without changing the exchange area for the heat removal from the reactor. Preliminary results of this study have been published<sup>2</sup>.

### EXPERIMENTAL

*Design.* The basic part of the apparatus is a vessel filled with water which mimics the reaction mixture. The vessel accommodates a stirrer, a thermocouple, and electric heating cells. The mixture

temperature is recorded by a computer in a closed loop. By means of control programs, the simulated reaction rate, degree of conversion, and heat production rate are calculated from the chosen rate equation based on the temperature data measured. The calculated amount of heat then is delivered into the simulation vessel by means of the computer-controlled electric heating. The heat is removed from the vessel by a real cooler divided into two sections: a closed section, mimicking the heat capacity of the reactor wall, and a flow-through section mimicking the cooling system volume. The cooling water flow rate is controlled by on-line opening and closing of a two-way valve.

*The apparatus* consists of two rectangular steel plate vessels sharing a wall for heat exchange. The larger vessel (23 dm<sup>3</sup>), accomodating a stirrer, represents the batch reactor (Fig. 1), the smaller vessel (10 dm<sup>3</sup>) is the cooler. Water serves both as the model reaction system and coolant; its volume in the reactor is 13 dm<sup>3</sup> and its level is so adjusted that the exchange area is 5.4 dm<sup>2</sup>. The vessel complex is insulated from the environment with a polystyrene board 10 cm thick.

*Heating.* The heating is provided by two electric heating coils submerged in the water. The voltage applied is obtained from a thyristor unit serving as the control signal amplifier. The heating control circuit comprises four resistors connected in series; their resistances are in the ratio 1 : 2 : 4 : 8. This resistor system forms the D/A converter or the conversion of the computer output signal to the thyristor unit control signal. Seventeen different power values in the 0 to 1 600 W range are obtainable by resistor combinations; these are set by the computer so that the amount of heat calculated by integration of the rate equation entered can be delivered to the reaction system. The accuracy of the heat delivery is estimated to be 5% relative.

*Cooler.* A closed cooling circuit is used. The cooler volume can be reduced by attaching polystyrene boards to the wall opposite to the heat exchange surface. In the cooler, a steel plate separates the flow-through section from the closed section; its position is adjustable. The cooling water circulation is forced by means of a centrifugal pump. The flow rate is measured with a rotameter and controlled manually by means of two throttle valves at the pump delivery and bypass. A thermostatted bath serves for controlling the entering coolant temperature.

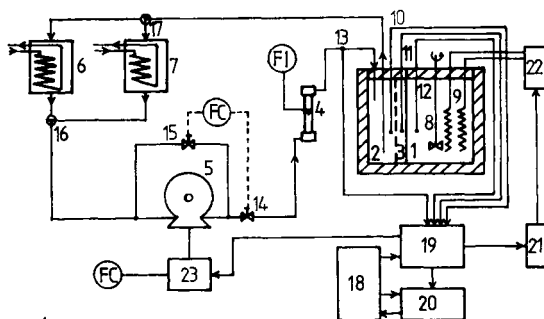


FIG. 1

Layout of the simulation apparatus. 1 reactor, 2 cooler (flow-through section), 3 cooler (closed section), 4 rotameter, 5 pump, 6, 7 thermostats, 8 stirrer, 9 heating coils, 10–13 thermocouples, 14, 15 throttle valves, 16, 17 three-way valves, 18 process control computer, 19 channel selector, 20 digital voltmeter, 21 D/A converter, 22 thyristor unit, 23 relay

*Temperature measurement.* The temperature of the water in the reactor and in the two cooler sections and the inlet temperature of the cooling water are measured with copper-constantan thermocouples with respect to a reference junction at 273.15 K. The time constant of the thermocouples is 4–5 s.

*Computer and logger units.* The temperature measurements, chemical reaction simulation and coolant flow rate control were accomplished by means of a Hewlett-Packard 3052A automated data acquisition system, comprising an HP 9835A process control computer, an HP 3495A scanner and an HP 3455A precision multimeter.

## THEORETICAL

### MATHEMATICAL MODEL OF THE SYSTEM

The model was set up for a perfectly stirred batch reactor in which an exothermic chemical reaction takes place, and for the flow-through cooler section perfectly mixed by the coolant.

The dynamic behaviour of the system is described by three first order ordinary differential equations derived from the mass and enthalpy balance of the reactor and the enthalpy balance of the cooler. The mass balance of the reactor is described by the equation

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

the enthalpy balance of the reactor is given by the equation

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

the parameters characterizing the heat exchange intensity for the reaction mixture–cooler system ( $A_R$ ) and for the reaction mixture–environment system ( $A_Z$ ) are

$$A_R = UP/(V\rho c_p)_R \quad (3)$$

and

$$A_Z = U_Z P_Z / (V\rho c_p)_R. \quad (4)$$

The enthalpy balance of the cooler can be written as

$$dT_C/dt = A_C(T - T_C) - C(T_C - T_{Ci}); \quad (5)$$

parameter  $A_C$  characterizing the heat exchange intensity for the cooler–reaction mixture system is given by the relation

$$A_C = UP/(V\rho c)_C \quad (6)$$

and parameter  $C = F_C/V_C$  is the inverse mean residence time of coolant in the cooler.

The reactor and cooler time constants are given by the relations

$$\tau_R = 1/(A_R + A_Z) \quad (7)$$

and

$$\tau_C = 1/(A_C + C). \quad (8)$$

The heat production rate for the chemical reaction simulation is calculated as

$$r_h = (V \rho c_p)_R \Delta T_{ad} (dx/dt). \quad (9)$$

The instantaneous degrees of conversion are obtained by integration of Eq. (1), the temperature of the model reaction mixture is measured with a thermocouple.

#### MODEL RATE EQUATION

The basis for the formulation<sup>3,4</sup> of the model rate equation was the highly exothermic oxidation of ethanol by hydrogen peroxide, homogeneously catalyzed by ferric ions<sup>5</sup>. The reaction enthalpy of this reaction is  $-275 \text{ kJ mol}^{-1}$  and the activation energy is  $90 \text{ kJ mol}^{-1}$ , both with respect to  $\text{H}_2\text{O}_2$ . The kinetics of this reaction can be described, with a sufficient accuracy, by a reaction of first order with respect to  $\text{H}_2\text{O}_2$ . The parameters of the equation were chosen such that unstable pseudostationary states occurred at the limiting attainable heat-transfer coefficient values. The adiabatic temperature increase of the reaction was chosen to be 130 K and the activation energy  $120 \text{ kJ mol}^{-1}$ . The rate equation

$$dx/dt = 2 \cdot 10^{15} \exp(-14433/T) (1-x) \quad (10)$$

was used.

#### CONTROL PROGRAM

The process control computer operates based on a real-time program, using two timing units. The first of them is used for the time measurement, the other makes it possible, within a preselected time interval  $\Delta t_h$ , to control the electric heating and to implement other functions of the program. The experiments can be performed with or without simulation of the chemical reaction, according to the user's choice.

In the main program, input data are read in and functions to be implemented are specified. The electric heating is also connected and disconnected by this program. After a preselected integral multiple of the interval  $\Delta t_h$ , subroutines for the various functions are called. After each disconnection of heating, the program waits for the start of a next  $\Delta t_h$  interval.

Subroutine MEASURE performs the thermocouple voltage measurements and the related temperature calculations. Subroutine POLY provides reaction mixture temperature data smoothing over a given curve segment, employing the multiple linear regression method on a polynomial. Subroutine DIFER calculates the reaction temperature derivative with respect to time and performs system identification. The decision block for the on-off temperature control is also contained in this subroutine. Subroutine COOL engages and disengages the coolant flow by means of an auxiliary quantity which acquires the value of 0 or 1 according to the conditions. Subroutine SELECT calculates, from the entered rate equation, the reaction rate, degree of conversion, heat production rate, and amount of heat to be produced within the next  $\Delta t_n$  interval. Subroutine TISK controls data printout on a chosen output device. Subroutine INIT from the system library of the computer ensures initialization of the logger and the timing units.

## RESULTS AND DISCUSSION

### DYNAMIC PROPERTIES OF COOLER

The dynamic properties of the cooler play an important part in the reactor control. The inertia of the cooling system arises from the accumulation of heat and coolant in the cooling coil or jacket and from the accumulation of heat in the reactor wall. Thus, the cooling system behaves like a system of two time-lag elements connected in series, where the response of the first element is significantly dependent on the coolant flow rate whereas that of the other element is independent of it. The simulation apparatus enables the behaviour of the cooling system to be imitated. The flow-through cooler section imitates the cooler volume, the closed section, the reaction wall (Fig. 1). The total cooler volume, the volumes of the two sections, and the resistance to heat transfer then can be varied by adjusting the plate separating the two cooler sections and by inserting polystyrene boards. Examples of study of the dynamic properties of the cooler are given in Table I; the results shown were derived from the transfer characteristics obtained as responses to the jump change in the temperature of entering coolant for various adaptations of the cooler.

The results illustrate the possibility of increasing the cooler inertia at a constant heat-transfer area. The volume ratio of the flow-through to the closed cooler sections can be varied without changing appreciably the heat transfer coefficient.

### EXAMPLES OF TEMPERATURE CONTROL SIMULATION

For the temperature control of reactors with exothermic reaction, the reactor contents have first to be heated to the desired temperature and then cooled for removing the reaction heat. Two problems thus must be solved.

1. The moment of switching from heating to cooling must be determined. This must be carried out in advance to counteract the cooler inertia.
2. The algorithm for keeping the reaction mixture temperature at the desired value must be used. The demands placed on the control algorithm are also dependent on the dynamic properties of the cooler.

Both the heat-transfer coefficient and the cooler inertia are involved in the treatment of this problem.

For illustrating the function of the equipment, the reaction temperature control was applied to a state which is unstable in the open control loop. A simple algorithm of a "relay with hysteresis" was employed for the computer feedback temperature control. The manipulated variable was the coolant flow rate  $F_C$ ; the flow was either completely open or completely shut.

The effect of cooler inertia on the temperature control in the unstable pseudostationary state was studied at two heat-transfer coefficient values and three  $V_C/F_C$  ratios.

The desired reaction temperature was reached adiabatically. Therefore, by the moment of switching from heating to cooling, the cooler contents had been heated nearly to the reaction mixture temperature. The results indicate that under these conditions the switching from heating to cooling must be accomplished before the reaction temperature attains the desired value.

The effect of the advance is shown in Figs 2–7. A comparison of Figs 2–4 demonstrates the effect of the cooler inertia on the requisite advance at heat-transfer coefficient values about  $800 \text{ W m}^{-2} \text{ K}^{-1}$ . The results document the fact that the advance has to be increased with increasing cooler inertia. The analogous dependence for heat transfer coefficient values about  $400 \text{ W m}^{-2} \text{ K}^{-1}$  is documented by Figs

TABLE I  
Cooler parameters for the various adaptation variants used

Variant No	$V_C$ $\text{dm}^3$	$V_W$ $\text{dm}^3$	$F_C$ $\text{cm}^3 \text{ s}^{-1}$	$C \cdot 10^3$ $\text{s}^{-1}$	$U$ $\text{W m}^{-2} \text{ K}^{-1}$	$\tau_R$ s	$\tau_C$ s
1	2.6	0.8	12.0	5.7	320	3 200	140
2	2.6	0.8	29.0	11	310	3 300	80
3	2.6	0.8	36.0	14	340	3 000	70
4	4.3	0.8	29.0	6.8	360	2 800	130
5	7.2	0.8	29.0	4.1	300	3 400	220
6	2.6	2.4	29.0	11	260	4 000	80
7	2.6	5.4	29.0	11	210	4 800	80
8	2.0	0	36.0	18	840	1 200	40
9	8.0	0	36.0	4.5	750	1 400	170

5–7. It follows from a comparison of Figs 2 and 5 that a decrease in the heat-transfer coefficient brings about a lowering in the pseudostationary reaction temperature. An additional consequence of the slower heat transfer is a slower response of the system to the activation of the coolant flow. With a low cooler inertia this slowing-down practically does not appear and the requisite advance in the switching from heating to cooling is the same for the two heat-transfer coefficient values (Figs 2 and 5). A comparison of Figs 3 and 6 or 4 and 7 shows that with increasing cooler inertia the requisite advance increases more at a poorer heat transfer.

The results also show that with the control algorithm used, the hazard of an uncontrolled temperature rise is greater at a higher cooler inertia or poorer heat transfer. While allowing for a successful temperature control at a low cooler inertia (Figs 2, 5) and high heat-transfer coefficient (Figs 2, 3), the algorithm of a relay with hysteresis fails if the cooler inertia is too large or the heat transfer is too poor.

### CONCLUSIONS

The results of the experiments indicate that the simulation reactor devised mimics well the behaviour of a reactor with exothermic reaction, thus making it possible for phenomena affecting the temperature control of chemical reactors to be studied in detail.

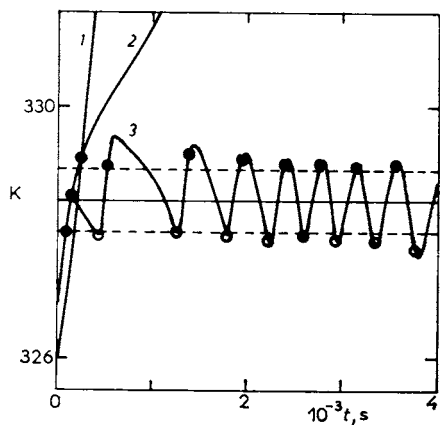


FIG. 2

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $U = 800 \text{ W m}^{-2} \text{ K}^{-1}$ ,  $V_C/F_C = 500 \text{ s}$ , advance: 1 0 K, 2 0.5 K, 3 1 K; ● cooling activation, ○ cooling deactivation

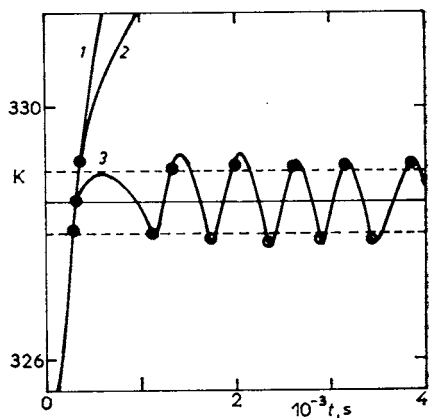


FIG. 3

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $V_C/F_C = 1000 \text{ s}$ , otherwise as in Fig. 2

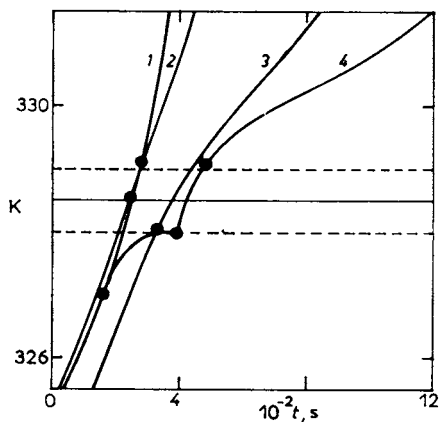


FIG. 4

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $V_C/F_C = 2000$  s, otherwise as in Fig. 2

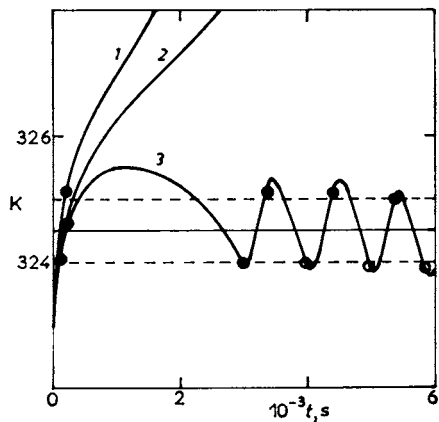


FIG. 5

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $U = 400 \text{ W m}^{-2} \text{ K}^{-1}$ , otherwise as in Fig. 2

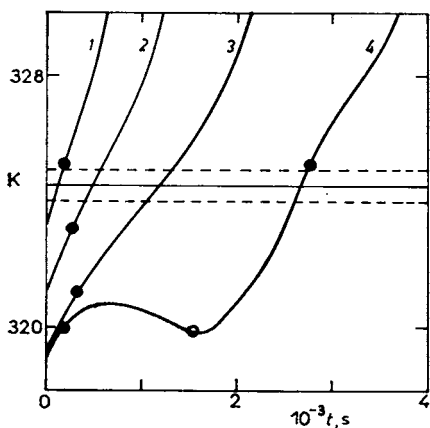


FIG. 6

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $U = 400 \text{ W m}^{-2} \text{ K}^{-1}$ ,  $V_C/F_C = 1000$  s, advance: 1 0 K, 2 2 K, 3 4 K, 4 5 K; ● cooling activation, ○ cooling deactivation

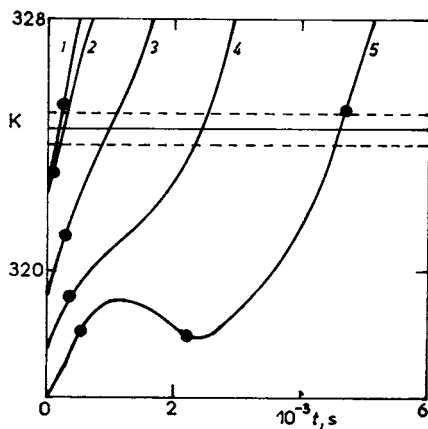


FIG. 7

Effect of the moment of switching from heating to cooling on the reaction temperature course;  $U = 400 \text{ W m}^{-2} \text{ K}^{-1}$ ,  $V_C/F_C = 2000$  s, advance: 1 0 K, 2 2 K, 3 4 K, 4 6 K, 5 7 K; ● cooling activation, ○ cooling deactivation



The analysis of the cooler's dynamic properties showed that these properties can be varied over a sufficiently wide span keeping the heat exchange area constant. The cooler inertia (time constant) can be varied by adjusting the ratio of the volume of the flow-through cooler section to the coolant flow rate at a practically constant heat-transfer coefficient value. This coefficient, and hence, the time constant of the reactor, can be changed by adjusting the volume of the closed cooler section.

These adjustments can be employed for the investigation of the effect of the cooler inertia on the regime stability and on the safety of the isothermal temperature control in the unstable pseudostationary state. The results give evidence that the simple relay control is only safe if the cooler inertia is low enough and the heat transfer from the reaction mixture to the cooler is sufficiently intense.

#### LIST OF SYMBOLS

$A$	parameter characterizing heat exchange intensity ( $s^{-1}$ )
$C$	$F_C/V_C$ , the inverse mean residence time of coolant in cooler ( $s^{-1}$ )
$c_{AO}$	initial concentration of key component ( $mol\ dm^{-3}$ )
$c_p$	specific heat ( $J\ g^{-1}\ K^{-1}$ )
$E$	activation energy ( $kJ\ mol^{-1}$ )
$F_C$	coolant flow rate ( $m^3\ s^{-1}$ )
$P$	heat exchange area ( $m^2$ )
$r$	reaction rate ( $mol\ dm^{-3}\ s^{-1}$ )
$r_h$	heat production rate ( $kJ\ s^{-1}$ )
$T$	reaction mixture temperature (K)
$T_C$	coolant temperature in cooler (K)
$T_{Ci}$	inlet coolant temperature (K)
$\Delta T_{ad}$	adiabatic temperature increase of reaction (K)
$t$	time (s)
$U$	heat-transfer coefficient ( $W\ m^{-2}\ K^{-1}$ )
$V$	volume ( $m^3$ )
$x$	degree of conversion
$\rho$	density ( $kg\ m^{-3}$ )
$\tau$	time constant (s)

#### Subscripts:

h	heat production
i	cooler inlet
R	reactor, reaction mixture
W	closed section of cooler

#### REFERENCES

1. Mukesh D., Cooper A. R.: *Ind. Eng. Chem. Fundam.* 22, 145 (1983).
2. Horák J., Beránek P.: *CHISA Natl. Conference*, High Tatras (Czechoslovakia) 1983.
3. Jiráček F., Horák J.: *This Journal* 47, 454 (1982).
4. Ali H., Bansal M. C., Alam S. S.: *Chem. Eng. World* 13 (3), 51 (1978).
5. Horák J., Jiráček F., Ježová L.: *This Journal* 48, 2627 (1983).

Translated by P. Adámek.