Transient State Simulation of a Distributed Parameter System:

Control of Hot Spot in a Packed-Bed Tubular Reactor

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In the chemical process industry many reactions are carried out in externally cooled packed-bed tubular reactors. If the reaction is highly exothermic, such as commercial hydrocracking (2), the reaction is likely to produce a hot spot at some point in the reactor where the temperature and rate of reaction increase almost instantly in spite of the external cooling (see curve A in Figure 1). The existence of a hot spot is generally very undesirable since it may influence production, product selectivity, catalyst life, or possibly even melt the reactor (2).

Paris (8) has proposed a scheme to eliminate the hot spot in an externally cooled packed-bed tubular reactor by dividing the reactor into different sections, each with a different rate of external cooling. The first few sections have a low rate of cooling to allow the reaction to start and to allow the temperature to reach a desired level. The next sections, where the reaction rate is very high, have a high rate of cooling to take up the heat of reaction so that the temperature will not rise further. The last sections again have a low rate of cooling so that the reaction will go to completion at the desired temperature.

To study the problem of hot-spot elimination in a packed-bed tubular reactor, Paris developed a reaction model, proposed a reactor design, and simulated both the steady and transient states of this reactor using numerical analysis on a digital computer (9).

Extensive amounts of work have been done with the simulation and control of lumped parameter systems, such as a continuous stirred-tank reactor, using both digital and analog equipment. The analysis of the distributed parameter system in the steady state has also been quite common with digital and analog equipment. However, an accurate simulation of the dynamic state of a distributed parameter system has required a large amount of time on a digital computer or an excessive amount of equipment on an analog computer. Paris found that the complete transient simulation of his reactor took about 130 min. on an IBM 705 computer. The analysis of a controller and control settings for his reactor at about 2 hr. a run on a digital computer would be exorbitant.

Since slight changes in the input parameters may greatly change the operating characteristics of any reactor, a control system is usually mandatory. The purpose of this paper is to show how a control system for a packed-bed tubular reactor may be simulated on conventional analog equipment using the technique of automatic iterative operation.

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ANALOG SIMULATION OF STEADY STATE

The reaction model used in this study is the same one used by Paris in his analysis (8, 9). Reactant A is converted to product B in an irreversible exothermic reaction

$$A \rightarrow B$$

Both A and B are gaseous and monomolecular, so there is no change in volume.

The reaction takes place in a packed-bed tubular reactor: a cylinder 4 ft. long and 2 in. wide, filled with porous spherical catalyst particles of ⅛-in. diam. As reactant A and an inert gas enter and flow through the reactor, species A diffuses into the pores of the catalyst to the available surface where the reaction occurs and species B is produced. The temperature of the reactor is controlled by a flow of coolant gas external and perpendicular to the reactor tube.

The following assumptions are made:

1. The pressure in the reactor is constant at 1 atm. The density, heat capacity, and viscosity of the gas are constant.
2. The catalyst particles are identical with constant heat capacities and have catalytic activity independent of temperature.
3. The reaction is highly exothermic with constant heat of reaction. Reaction takes place only when adsorbed on the surface of the catalyst. The reaction is rate limiting

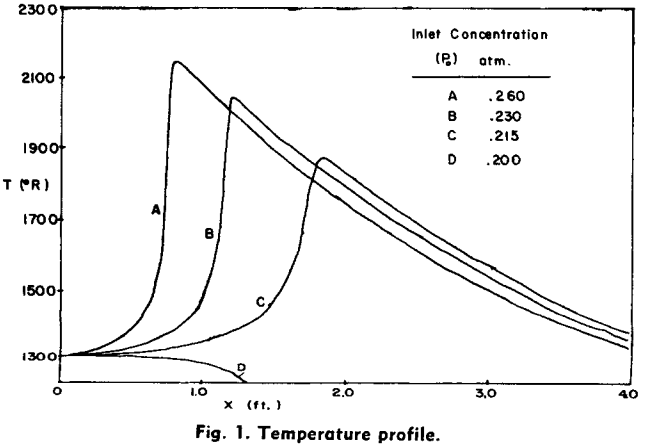

Fig. 1. Temperature profile.

with overall apparent rate being first order with respect to the partial pressure of A and with an Arrhenius type dependence on temperature. Rate $= k_0 p e^{(-E/RT)}$.

4. The reactor is of the plug-flow type.

A mass balance and an energy balance along the reactor yield the following partial differential equations governing the reaction:

$$\frac{Q}{P}\left(\frac{\partial p}{\partial x}+\frac{1}{u}\frac{\partial p}{dt}\right)+(1-\epsilon)\left(\epsilon p\frac{\tilde{e}}{p}\right)\frac{\partial p}{\partial t}= -(1-\epsilon)p_p\,\sigma k_0 p e^{-E/RT} \quad (1)$$

$$Qc\left(\frac{\partial T}{\partial x}+\frac{1\partial T}{u\partial t}\right)+(1-\epsilon)(\epsilon p\,\tilde{\tilde{c}}\,p+c_p\,p_p)\frac{\partial T}{\partial t} =\frac{4h_e}{d_t}(T_c-T)+(1-\epsilon)\,\sigma p_p\,k_0 p e^{-E/RT}(-\Delta H) \quad (2)$$

These will henceforth be abbreviated to

$$A\left(\frac{\partial p}{\partial x}+\frac{1}{u}\frac{\partial p}{\partial t}\right)+F\frac{\partial p}{\partial t}=-Bpk \quad (3)$$

$$C\left(\frac{\partial T}{\partial x}+\frac{1}{u}\frac{\partial T}{\partial t}\right)+K\frac{\partial T}{\partial t}=D(T_c-T)+Bpk(-\Delta H) \quad (4)$$

$$k=k_0 e^{-E/RT} \quad (5)$$

For the steady state solution the derivative terms with respect to time are equal to zero, so Equations (3) and (4) reduce to a set of ordinary differential equations:

$$\frac{Adp}{dx}=-Bpk \quad (6)$$

$$\frac{CdT}{dx}=D(T_c-T)+B(-\Delta H)pk \quad (7)$$

$$k=k_0\epsilon^{-E/RT} \quad (8)$$

which may be solved directly on the analog computer.

Figure 1 shows steady state temperature profiles and illustrates the phenomenon known as parametric sensitivity. Small changes in the parameters of the system can change the position and severity of the hot spot and thus greatly affect reactant conversion. Figure 1 shows only the effect of inlet concentration, although catalyst activity, inlet temperature, and heat transfer also affect reactor performance in a similar manner.

Curve A of Figure 1 illustrates the typical behavior of a hot spot in a tubular reactor. In the first 0.5 to 1 ft. of the reactor the reaction rate is low. Heat produced in the reaction is slightly greater than the heat removed by external cooling, so the temperature starts to rise. As the temperature rises the rate increases until it "catches fire." At this point the heat generated greatly exceeds the heat removed, producing an almost vertical temperature profile, very high temperature, and a very high rate of reaction. Within a short space the high reaction rate consumes almost all of the reactant and the rate drops nearly to zero. External cooling then gradually lowers the temperature. In curve D of Figure 1 the temperature does not rise sufficiently high or fast, resulting in very little conversion of reactant into product.

Paris found that the hot spot could be eliminated and a high degree of conversion maintained if he divided the reactor into five sections, each with a different rate of external cooling (Figure 2). Heat transfer coefficients in each section were adjusted in order to approximate a temperature profile that rose linearly from 1,300° to 1,500°R. in the first half of the reactor, and remained constant for the remainder of the reactor. The analog simulation of the present study required the use of high-speed electronic comparators to change the heat transfer coefficient along the reactor length. Since only three comparators were available, the final half of the reactor could not be studied in repetitive operation.

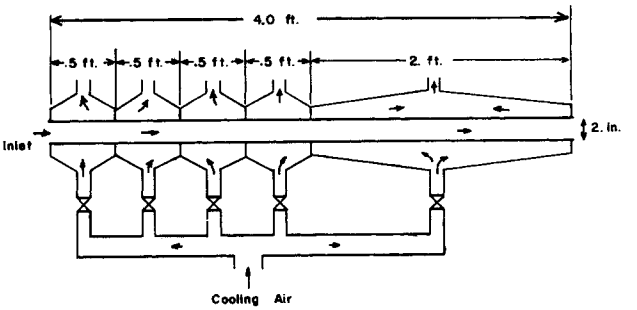

Fig. 2. Reactor and cooling system.

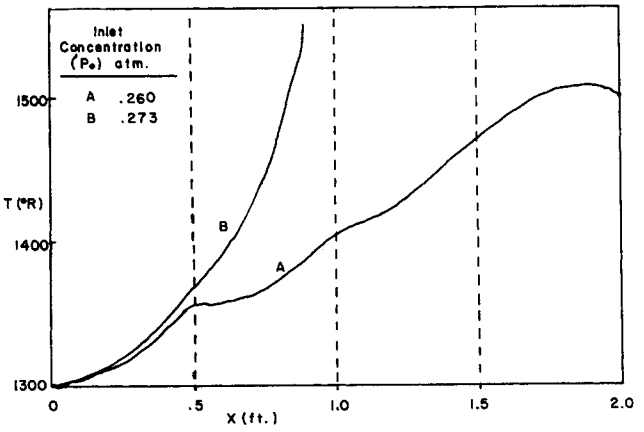

Fig. 3. Temperature profiles with sectional cooling.

Temperature profiles for the first half of the reactor with sectional cooling are shown in Figure 3. Even in the final steady state design with sectional cooling, the system remained sensitive to small changes in inlet conditions. Figure 3 shows the effect of a 5% increase in inlet concentration and indicates the development of a hot spot. Clearly, some means must be found for the detection and control of a hot spot caused by variations in inlet conditions.

DYNAMIC SIMULATION SCHEME

The simulation of a distributed parameter system in the transient state requires the solution of partial differential equations. In the case of the tubular reactor, the equations are the mass balance and energy balance of the reactor [Equations (3), (4), and (5)]. An analog computer solution of partial differential equations usually involves a transformation of each partial differential equation into a set of ordinary differential equations since the analog computer can only integrate with respect to one variable (*1*). The technique used in this study to transform the partial differential equations to ordinary differential equations is related to the method of characteristics (3). By this method it may be shown that

$$\frac{1}{u}\frac{dp}{dt}=\frac{dp}{dx}+\frac{\partial p}{\partial x}+\frac{1}{u}\frac{\partial p}{\partial t} \quad (9)$$

and

$$\frac{1}{u}\frac{dT}{dt} = \frac{dT}{dx} = \frac{\partial T}{\partial x} + \frac{1}{u}\frac{\partial T}{\partial t} \qquad (10)$$

where u is the velocity of gas flow in the tubular reactor.

Applying these transformations to Equations (3) and (4) and considering the remaining partial terms as negligible, we get the following:

$$A\frac{dp}{dx} = -Bpk \qquad (11)$$

$$C\frac{dt}{dx} = D(T_c - T) + Bpk(-\Delta H) \qquad (12)$$

Equations (11), (5), and (12) may then be integrated directly on the analog computer. The path of integration will follow the characteristic lines on the space-time domain of Figure 4. These lines are the parallel lines of slope equal to $1/u$ in Figure 4.

Another way of looking at the method of characteristics is to consider a small plug of fluid that enters the tubular reactor at time $t = 0$ and length $x = 0$ (5). As this plug of fluid moves down the reactor, it will follow the path of the first dark characteristic line of Figure 4. As time goes on it moves down the length of the reactor at velocity u until the plug leaves the reactor at time equal to one residence time. The ordinary differential equations resulting from the characteristic transformation of the partial differential equations governing the tubular reactor are equivalent to the ordinary differential equations that govern a batch reactor. The method of characteristics is then equivalent to approximating the tubular reactor as a series of small batch reactors flowing down the length of the reactor at a velocity equal to u.

The method of characteristics may be correctly applied to the packed-bed tubular reactor only if the partial terms corresponding to the heat capacity and the matter residence are negligible [terms $F(\partial p/\partial t)$ and $K(\partial T/\partial t)$ in Equations (3) and (4)]. Paris found that the partial of pressure with respect to time was nearly negligible but that the partial of temperature with respect to time was quite significant. He reported residence times as follows:

Flow residence time	0.39 sec.
Concentration residence time	0.68 sec.
Heat residence time	6.37 min.

Both residence times are larger than the residence time for the flowing gas alone due to the capacitance of the catalyst. Since the matter capacitance is small compared to the gas at large, the matter or concentration residence

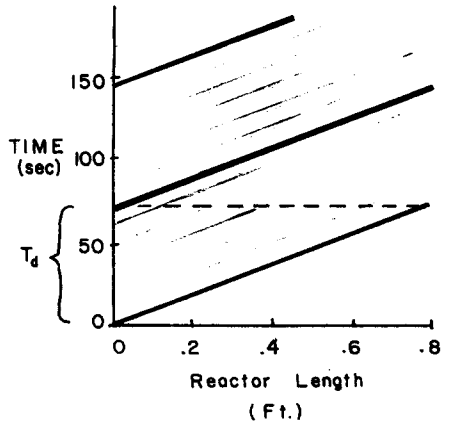

Fig. 4. Space-time domain

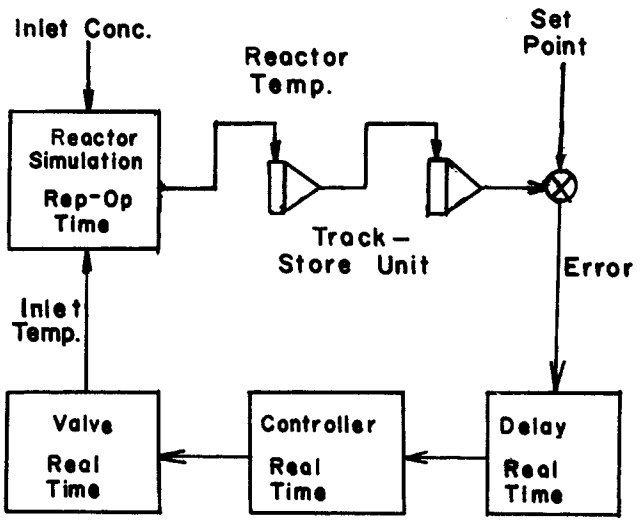

Fig. 5. Block diagram of control system.

time is nearly equal to the true residence time. However the heat capacity of the catalyst pellets is quite large when compared to the heat capacity of the flowing gas. Consequently, a disturbance in temperature in the reactor moves down the length of the reactor much slower than the actual flow of gas. While the gas flows at a velocity u = 10.2 ft./sec., a temperature disturbance moves at a velocity v = 0.011 ft./sec. (see Figures 32, 34, and 36, pp. 140-143, of reference 8). Since reaction rate is a function of catalyst pellet temperature, the method of characteristics has been modified so that characteristic lines on the space-time domain follow the heat velocity (defined by heat residence time) instead of the gas velocity. This means that the characteristic lines in Figure 4 are at a slope $1/v$ instead of $1/u$.

A block diagram of the control system proposed and simulated is shown in Figure 5. A PACE TR-48 analog computer was used to simulate the reactor in rep-op time while a PACE TR-10 analog computer simulated the delay, controller, and valve in real time.

The technique of operating one part of a circuit at high speed rep-op time while slowly changing some of the parameters in slow speed real time is known as iterative operation (6). A rep-op cycle traces out a temperature profile along one of the characteristic lines of the space-time domain. Two integrators were modified to form a track-store unit. The tracking amplifier (reset coil patched to operate bus and operate coil patched to reset bus) follows the temperature along the reactor length when the computer is in "operate," and stores its final value when the computer is in "reset." The storage amplifier (patched normally) stores in "operate" and tracks in "reset." The track-store combination puts out a semicontinuous (small steps) output which is the final temperature at the end of the rep-op cycle. The length of the rep-op cycle determines the position of the final temperature or sensing temperature to be used by the controller.

A real time delay is needed to allow filling in of the space between the dark characteristic lines of the space-time domain. Without a time delay, each rep-op cycle would start a new characteristic line at the point where the previous characteristic line ended, that is, rep-op cycles would follow the dark lines in the space-time domain. The time delay

$$T_d = \frac{x}{v} = \text{(residence time of temperature disturbance from inlet up to point of sensing)}$$

allows additional segments between the dark lines to be considered so that the output from the track-store unit will be more nearly continuous. This time delay was simulated with a second-order Padé approximation (*1*) on the PACE TR-10 in real time.

The controller simulated was of the proportional-integral type with the conventional transfer function (*4*):

$$\frac{\text{Output}}{\text{Input}} = \text{Gain}\ (1 + \tau_I s)$$

A first-order real time filter was used to smooth out the small steps in the output of the track-store unit and to smooth the irregularities of the Padé approximation. A first-order lag corresponds to the response of a control valve or of the temperature-sensing device, and is compatible with hardware that would be used if the system were actually built.

The philosophy of the control system for the tubular reactor is to maintain high conversion of reactant into product, without the appearance of a hot spot. The parameters most likely to change conditions in the reactor are inlet temperature, inlet concentration, and catalyst activity. A temperature controller could easily be used to control inlet temperature to the reactor, however it is impossible to control catalyst activity and impractical to control inlet concentration, especially if the feed to the reactor is the product from some other operation. The control scheme selected was to place a temperature-sensing device at a point in the reactor either at or before the place of highest steady state reaction rate. This temperature sensing would detect the tendency of a developing hot spot by a rise in temperature, and would cause a corresponding decrease in inlet temperature as a result of the controller.

SIMULATION RESULTS

The two analog computers were programmed according to the Appendix, and the control system was simulated. Figures 6 and 8 to 11 show the results of a step input consisting of a 5% increase in inlet concentration. The temperature control point in each case was 0.8 ft. from the inlet of the reactor in the second cooling section.

Figure 6 shows a typical response to a step input with and without control. The units of gain in all figures are degree change in inlet temperature per degree change in set point temperature. Considerable noise can be seen in the curves even after the first-order lag, confirming the need for the filter. The ripples in the curves between the step input and the response at one time delay later are

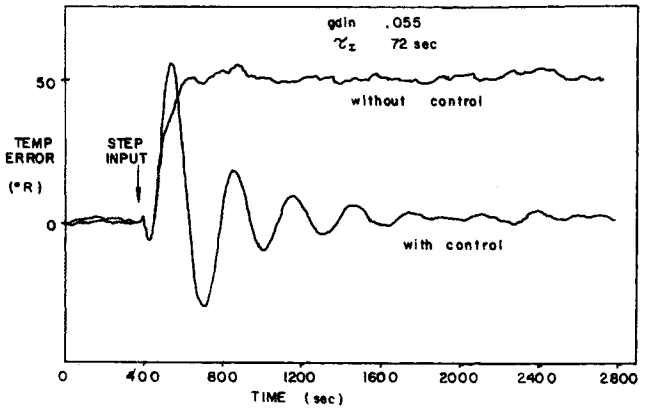

Fig. 6. Typical temperature response.

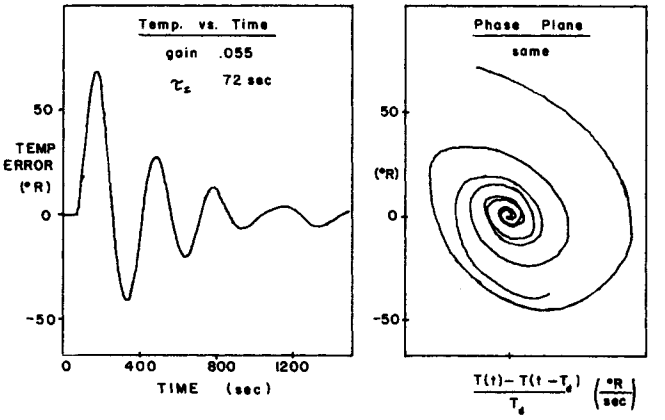

Fig. 7. Temperature response with phase plot.

characteristic of the second-order Padé approximation. It should be noted that the initial rise in temperature is greater with control than without control. This is due to response of the controller to the small negative ripple preceding the initial rise.

Figure 7 shows a temperature versus time curve and its corresponding phase plane. The phase plane shows two responses, one for a 5% positive increase in inlet concentration and the other for a 5% negative increase in inlet concentration.

Figure 8 shows the temperature response with the controller set according to the rules of Ziegler-Nichols (*4*). The response is somewhat conservative as is usually expected with Ziegler-Nichols control settings. Improved response was obtained in Figure 8 by decreasing the integral (reset) time.

Figures 9 and 10 show the effect of changing gain at constant reset time and of changing reset time at constant gain. In each figure the initial response with control is greater than the corresponding response without control due to the negative ripple of the Padé approximation. Considerable noise can be seen in the steady state profiles even after application of the first-order filter.

Figure 11 shows a direct analog computer plot and a plot calculated and directed by a digital computer for identical inputs and control settings for the proposed control system. The two plots are almost identical, tending to confirm the accuracy and value of the analog computer for simulation work. The analog plot has a slightly higher initial rise and is somewhat more oscillatory, probably due to the Padé approximation.

CONCLUSION

The steady and transient states of a packed-bed tubular reactor have been studied on an analog computer. The phenomenon of parametric sensitivity was demonstrated and the idea of Paris, to divide the reactor into different cooling sections, was shown to eliminate the hot spot while maintaining high conversion.

A control system was devised for the tubular reactor to ensure proper operation of the reactor regardless of small changes in inlet conditions. Using the technique of iterative operation and the method of characteristics, the control system was simulated and values of control parameters were selected.

The reactor model and this simulation are uncertain in some areas. In this and in the study by Paris, axial diffusion was neglected. The presence of significant axial diffusion would change both the steady and transient states, possibly producing multiple steady state solutions. The param-

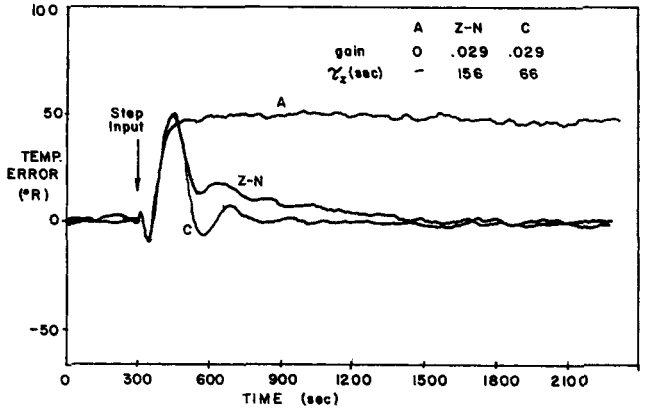

Fig. 8. Temperature response with Ziegler-Nichols control settings.

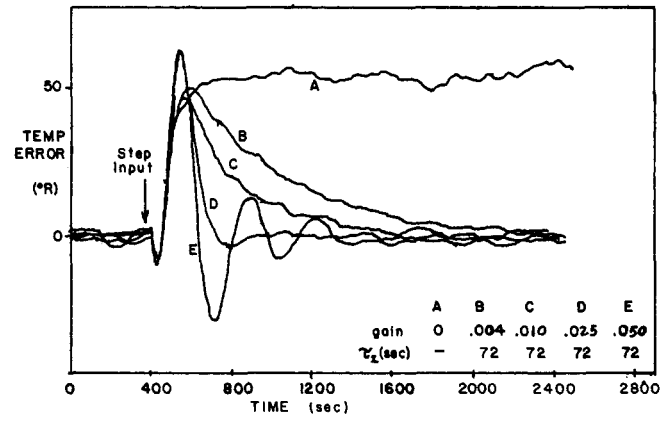

Fig. 9. Temperature response, effect of gain.

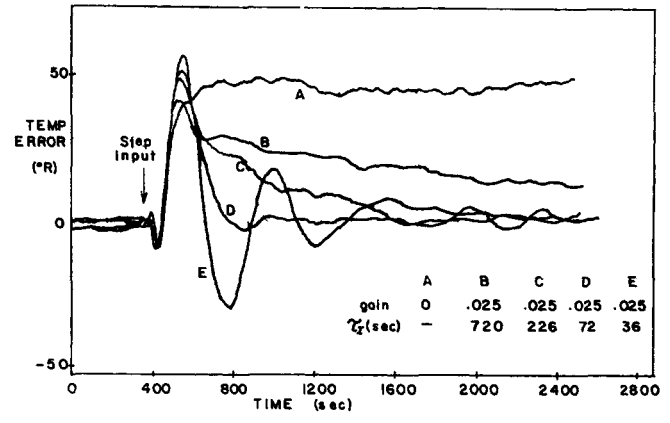

Fig. 10. Temperature response, effect of reset time.

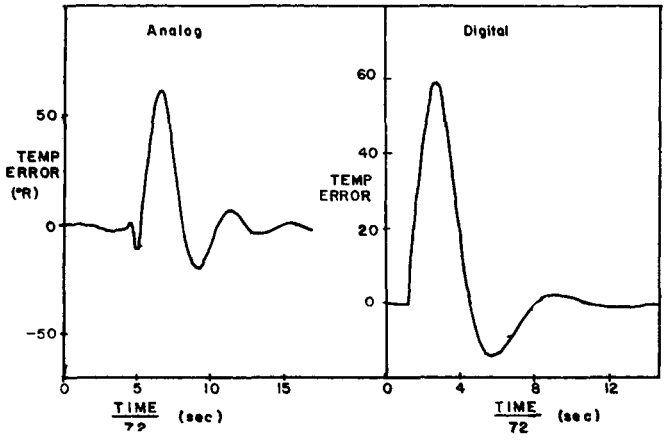

Fig. 11. Temperature response plots by analog and digital computers.

eters of the control system are highly dependent upon the value of the heat residence time. Before a control system is actually built this value should be confirmed experimentally.

NOTATION

A = reactant species
A = term in differential equation
B = product species
B = term in differential equation
C = term in differential equation
$\tilde{c}$ = molar heat capacity of gas stream, 36 B.t.u./(mole)(°R.)
c_p = heat capacity of pellets, 0.25 B.t.u./(lb.)(°F.)
D = term in differential equation
d_t = diameter of tube, 2 in.
E/R = activation energy/gas constant, 22,450°R.
F = term in differential equation
$(-\Delta H)$ = heat of reaction, 150,000 B.t.u./mole
h_e = overall effective heat transfer coefficient, 5.75 B.t.u./(sq.ft.)(hr.)(°F.)
K = term in differential equation
k_0 = reaction rate constant, 1.9 mole/(atm.)(hr.)(sq. ft.)
k = $k_0\exp(-E/RT)$
L = reactor length, 4 ft.
p = total pressure, 1 atm.
p = partial pressure of reactant
p_0 = inlet partial pressure, 0.26 atm.
Q = gas flow rate, total tube section, 12.5 mole/(sq. ft.)(hr.)
s = used in Laplace transforms, sec.$^{-1}$
t = time, sec.
T = temperature, °R.
T_c = coolant temperature, 800°R.
T_d = delay time, 72 sec.
T_0 = inlet temperature, 1,300°R.
u = average axial velocity of gas, 10.1 ft./sec.
v = average axial velocity of temperature disturbance, 0.011 ft./sec.
x, X = axial coordinate, ft.

Greek Letters

ϵ = bed porosity, 0.35
ϵ_p = particle porosity, 0.40
$\tilde{\rho}$ = molar density of gas, 9.78 × 16^{-4} mole/cu.ft.
ρ_p = density of pellets, 80 lb./cu.ft.
σ = surface area of pellets, 8 × 10^5 cu.ft./lb.
τ_I = integral time constant of controller
τ = first-order filter time constant

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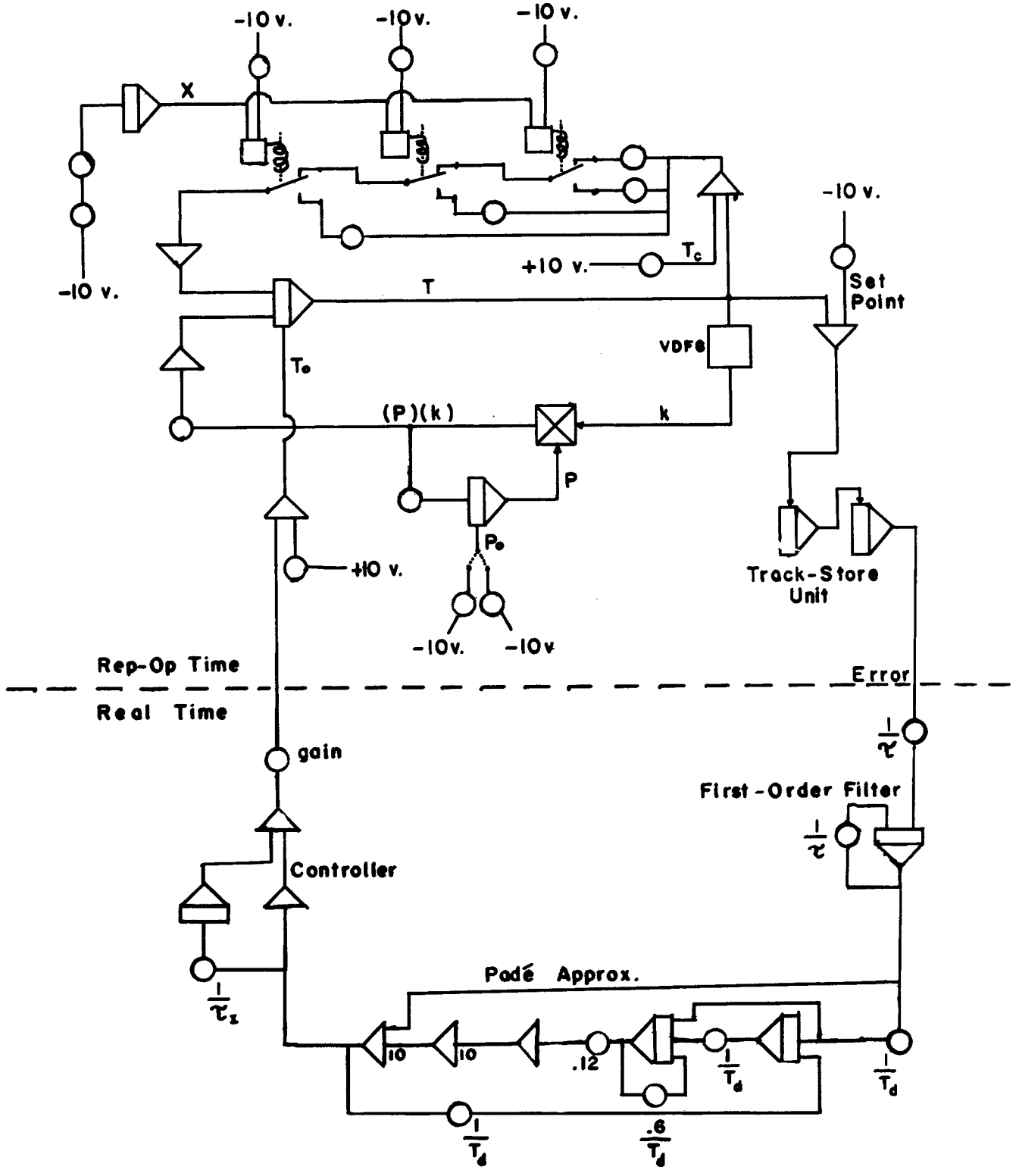

Fig. 12. Analog computer program.

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APPENDIX: ANALOG COMPUTER PROGRAM

Scaled equations:

$$\frac{d[3.5p]}{dx} = -\left(\frac{B}{1.7 \times 10^5 A}\right)[3.5p]\,[1.7 \times 10^5 k]\,[3.5p_0] = 0.91$$

$$\frac{d[0.001(T-1100)]}{dx} = \left(\frac{D}{C}\right)\{[0.001(800-1100)]$$

$$- [0.001(T - 1100)]\} + \left(\frac{0.001\,B(-\Delta H)}{3.5(1.7 \times 10^5\,C)}\right)$$

$$[3.5p]\ [1.7 \times 10^5\,k]$$

$$[1.7 \times 10^5\,k] = f\,[0.001(T - 1100)]$$

$$[0.001(T_0 - 1100)] = 0.200$$

Settings of variable diode function generator (Arrhenius approximation):

$[0.001(T - 1100)]$	$[1.7 \times 10^5\,k]$
0.0	0.0006
0.1	0.0024
0.2	0.010
0.3	0.035
0.4	0.103
0.5	0.258
0.6	0.595
0.7	1.250
0.8	↑
0.9	overload (7)
1.0	↓

Program details (see Figure 12).

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Behavior of Gas Bubbles in Fluidized Beds

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Apparently it is generally accepted that, of the total volumetric flow of gas upward through a fluidized bed of solid particles, only a small fraction stays in the particulate or emulsion phase where most of the solid particles are. The superficial gas velocity in this part of the bed is nearly the same as the superficial velocity at minimum fluidization u_{MF}, which is usually much smaller than the superficial velocity u corresponding to the total fluid flow rate. The excess of gas, corresponding to $u - u_{MF}$, flows through the bed as gas pockets or "bubbles."

Each of these cavities contains only a small mass of the solid phase and, for a fluidized catalytic reactor, the extent of contact between the solid catalyst and most of the fluid stream depends on the circulation of fluid in and out of the gas bubbles. Gas which does not escape the bubbles on their way through the catalyst mass will have by-passed the catalyst and will not have reacted. Thus, for the rational design of a fluidized reactor it is essential to know how rapidly gas flows between bubbles and emulsion and by what mechanism such exchange occurs.

PREVIOUS WORK

The most promising calculations of fluid flow patterns around a rising gas bubble in a fluidized bed are those of Davidson and Harrison (*6*) who conclude that the velocities of the solid particles around a bubble can be described by a velocity potential. Indeed, rising velocities of bubbles calculated on this basis were found to agree within about 10% with observed velocities. The data covered a variety of solid-particle sizes and densities and a wide range of bubble volumes. Their formula is

$$U_B = 0.71\,(g_L^3\,V_B)^{1/6} \qquad (1)$$

which closely resembles an earlier formula for the rising velocity of a large gas bubble in a liquid owing to Davies and Taylor (*8*). For two-dimensional gas bubbles in liquids contained between large flat plates the measurements of Collins (*13*) lead to

$$U_B = 0.66\,(g_L^2\,V_B/h)^{1/4} \qquad (2)$$

where h is the distance between plates.

With such surprising success in their application of potential flow methods to the bubble motion in fluidized beds, Davidson and Harrison went on to compute the relative movement of gas through the emulsion phase around a gas-filled cavity. They used the Darcy Law for this purpose, concluding that the pressure profile in the emulsion phase was represented by the Laplace equation with a uniform pressure on the surface of the cavity. In this way both gas and solid particle streamlines could be found around a cavity of assumed shape.

Figure 1 shows a few of their calculated gas streamlines around an assumed spherical cavity for the practically

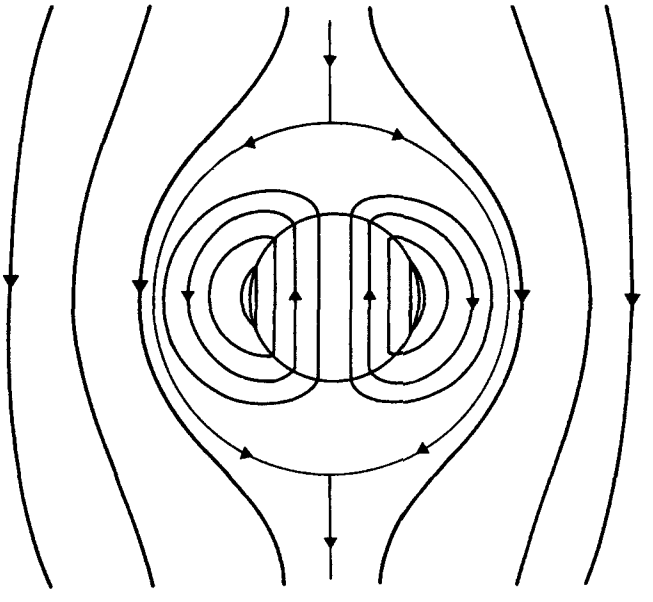

Fig. 1. Theoretically calculated gas-flow streamlines around spherical bubble in fluidized bed (after Davidson and Harrison, *loc. cit.*).

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