An Experimental Study of Oscillating Reactors

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The hydrolysis of acetyl chloride was studied in a laboratory reactor designed to act as a chemical oscillator. The observed oscillating outputs are in fairly good agreement with the numerical solutions of the stirred tank reactor equations and the approximate analytical solutions published previously.

It has been shown that the theoretical equations normally used to describe the operation of a continuous stirred tank reactor (CSTR) predict that for certain ranges of the system parameters, the reactor will generate periodic outputs even though the inputs are maintained constant (*1, 4, 5* to *8, 10*). In fact, these theoretical studies indicate that in some situations the time average conversions obtained from an oscillating reactor can be much higher (approximately 20 to 40%) than the steady state conversion (*5*) and that the profit obtained from a chemical oscillator can be greater than the optimum steady state design value (*6*). Since it is always dangerous to rely on theoretical predictions unless there is some experimental evidence which indicates that the theory is valid, it is necessary to make certain that the CSTR equations properly describe the reactor behavior when the system is generating oscillations.

Some experimental investigations of the dynamics of stirred tank reactors have been discussed in the literature (*9, 14*). However, the primary emphasis in these studies was to compare the theoretical and experimental step or impulse response of stable systems. It is well known that the CSTR equations predict an asymptotic approach to a steady state operating condition over wide ranges of the system parameters, but almost any other theoretical model also will exhibit this kind of behavior. In addition, the linearized CSTR equations will give almost exactly the same results as numerical solutions of the nonlinear equations for small changes in the system inputs. Therefore, it should not be surprising that an adequate agreement was obtained between the theory and the transient experiments.

This kind of experimental evidence does not provide a sound basis for the predictions of stable oscillating outputs which can be obtained from the CSTR model and which depend, to a large extent, on the nonlinear character of the equations. Hoffman (*8*) showed that the simple oscillator criteria he developed using a linear stability analysis could be used to forecast the existence of oscillations for the decomposition of hydrogen peroxide in an acetic acid solution with ferric ions as a catalyst, but he did not report a comparison between his model and the data.* Thus, an experimental study of oscillating reactors was undertaken in order to provide additional verification of the simple CSTR theory and to test some of the analytical solutions which have been published recently.

REACTOR MODEL

The hydrolysis of acetyl chloride was the reaction chosen for the investigation:

$$\underset{\text{acetyl chloride}}{CH_3COCl} + \underset{\text{water}}{H_2O} \rightarrow \underset{\text{acetic acid}}{CH_3COOH} + \underset{\text{hydrochloric acid}}{HCL} \qquad (1)$$

Providing that acetone is added to the reacting mixture, it is a homogeneous, liquid phase, exothermic reaction. The kinetics in acetone-water solvents have been studied by several investigators. Zimmerman and Yuan (*15*) made measurements in the range of 1 to 5% (by volume) water in acetone and postulated a trimolecular reaction mechanism. Swain and Scott (*12*) measured the reaction rate of a 25% (by volume) water solution at three different temperatures and calculated an activation energy for a pseudo first-order reaction. Cairns and Prausnitz (*2*) studied 10 to 25% water mixtures over a broad temperature range and proposed that the reaction was second order (first order with respect to water and first order with respect to acetyl chloride) but that the rate was strongly dependent on the dielectric constant of the solution. The results of Cairns and Prausnitz were used both by Tinkler (*13*) and Simpkins (*11*) in dynamic studies of tubular reactors and were found to give good predictions of the experimental data. Hence, we will consider that the reaction rate is second order.

If we assume that the physical properties of the mixture are independent of composition and temperature, that the only heat transfer is to the cooling coil in the reactor, that the arithmetic mean is a valid approximation of the log-mean driving force across the coil, and that the feed rate of each component to the reactor is maintained constant so that the stoichiometric equation can be used to relate the reacting components, then the material and energy balances can be written as

$$V\frac{dA}{dt} = q(A_f - A) - k\ V\ A\ (B_f - A_f + A) \qquad (2)$$

$$V\,C_P\rho\frac{dT}{dt} = q\,C_P\rho(T_f - T)$$
$$+ (-\Delta H)\ k\ VA\ \ (B_f - A_f + A) - \frac{UA_cKq_c}{(1 + Kq_c)}(T - T_c) \qquad (3)$$

where

$$k = k_0 \exp\ (-E/RT), \quad K = \frac{2C_{Pc}\rho_c}{UA_c} \qquad (4)$$

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* A recent paper [Bush, S. F., *Proc. Roy. Soc.*, A309, 1 (1969)] presents some experimental data for self-generated oscillations observed during the chlorination of methyl chloride in a CSTR.

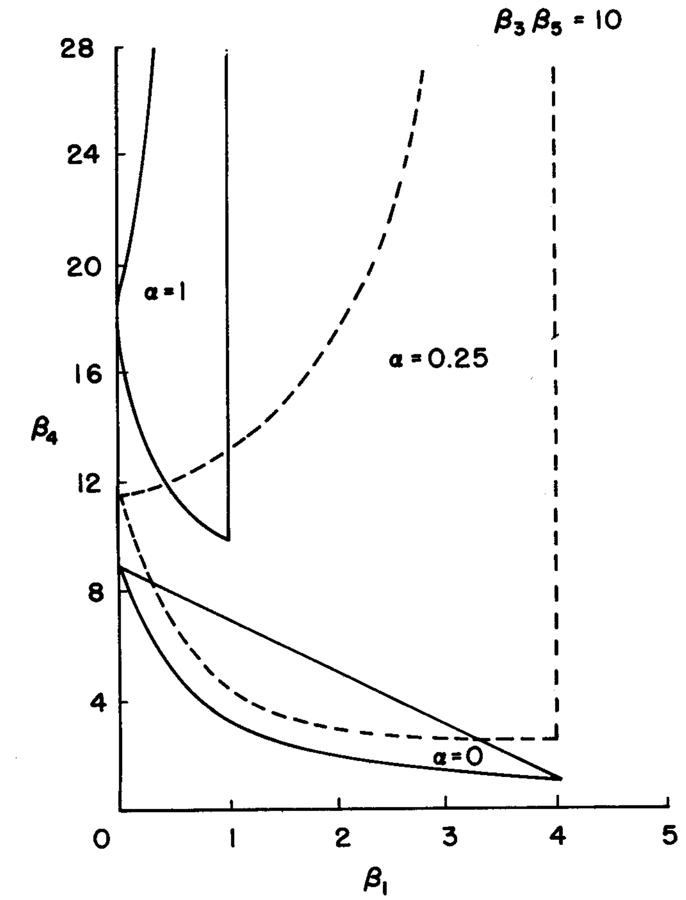

Fig. 1. Regions of oscillatory operation.

At steady state conditions, the accumulation terms are equal to zero. Then, Equation (2) can be solved for the steady state reactant composition A_s in terms of temperature and this result used to eliminate composition from the energy equation. After some manipulation, the result is

$$\left[q\,C_{P\rho} + \frac{UA_cKq_c}{1+Kq_c}\right]T_s - \left[q\,C_{P\rho}\,T_f + \frac{UA_cKq_cT_c}{1+Kq_c}\right] = \frac{1}{2}\,(-\Delta H)q\,\,(A_f + B_f + q/k_sV)\left[1 - \sqrt{1 + \frac{4A_fq/k_sV}{(B_f - A_f + q/k_sV)^2}}\,\right] \qquad (5)$$

It is not possible to solve this equation explicitly for the steady state temperature, since it is a transcendental equation. However, as is well known, the solutions can be found by plotting the left- and right-hand sides of the equation against temperature and by determining the intersections of the two curves. In order to keep our experiments as simple as possible, we always selected the design parameters in such a way that there was only one possible steady state operating condition. Also, we used the expression above to obtain new estimates of the reaction rate constant k, as will be discussed later.

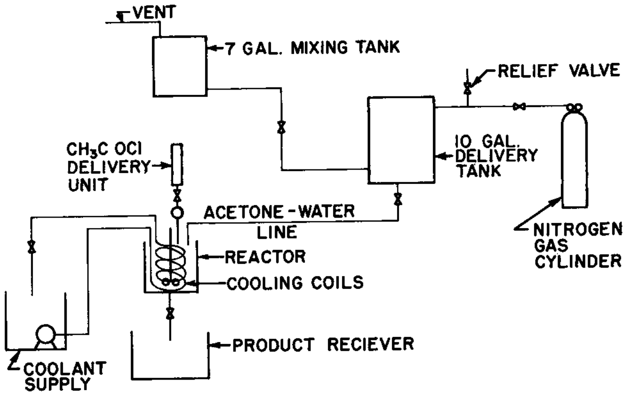

Fig. 2. Experimental equipment.

OSCILLATOR CRITERIA

Since we want the reactor to generate periodic outputs, we need to design the system so that the steady state solution of the mass and energy balance equations is unstable. In fact, this condition is almost sufficient to ensure oscillatory operation because it has previously been shown that the system is bounded and that all trajectories originating on the bounds must be directed toward the interior of the phase plane (*4, 6*). If we let

$$x_1 = -(A - A_s)/A_s, \quad x_2 = -(T - T_s)/T_s,$$

$$\beta_0 = (B_f - A_f)/A_s, \quad \beta_1 = k_sVA_s/q, \quad \beta_3 = E/RT_s,$$

$$\beta_4 = 1 + \frac{UA_cKq_c}{qC_{P\rho}(1 + Kq_c)} \quad \beta_5 = \frac{(-\Delta H)k_sVA_s^2}{qC_{P\rho}T_s}$$

$$\alpha = \frac{B_f - A_f}{A_f} \quad \tau = \frac{qt}{v} \qquad (6)$$

then the dynamic equations can be put into the form

$$\frac{dx_1}{d\tau} = -x_1 - \beta_1(1 + \beta_0) + \beta_1\left[(1 - x_1)^2 + \beta_0(1 - x_1)\right]e^{\beta_3x_2/(x_2-1)} \qquad (7)$$

$$\frac{dx_2}{d\tau} = \beta_4\,x_2 + \beta_5(1 + \beta_0) - \beta_5\left[(1 - x_1)^2 + \beta_0(1 - x_1)\right]e^{\beta_3x_2/(x_2-1)} \qquad (8)$$

This transformation merely translates the coordinate system so that the steady state operating point is at the origin.

After linearizing the equations, determining the characteristic roots, and requiring that the singular point must be an unstable focus or node, we find that the criteria for oscillatory operation are

$$1 + \beta_1\,(2 + \beta_0) + \beta_4 < \beta_3\,\beta_5(1 + \beta_0) < \beta_4\,[1 + \beta_1\,(2 + \beta_0)] \qquad (9)$$

Some algebraic manipulation of the quantities defined by Equation (6) leads to the expression

$$\beta_0 = \frac{\alpha(1 + \beta_1)}{1 - \beta_1} \qquad (10)$$

This result can be used to eliminate β_0 from the oscillator criteria given above, and then a set of parametric plots can be made which indicate regions where the system might have periodic outputs. An example showing the closed regions corresponding to oscillatory operation for $\beta_3\,\beta_5 = 10$ and three values of α is presented in Figure 1. This kind of a plot is equivalent to Hoffman's (*8*), except that it includes the possibility of having feed mixtures which are not in stoichiometric proportions. The oscillator region indicated on the graph is applicable only to cases where there is a single steady state operating point. If multiple steady state solutions are possible, then a more complete analysis is required (4).

APPROXIMATE ANALYTICAL SOLUTION

An approximate analytical solution for the periodic outputs generated by the reactor can be developed by following the procedure given in an earlier paper (6). The nonlinear functions on the right-hand sides of the dimensionless state equations, Equations (7) and (8), are expanded in a Taylor series about the steady state solution of interest, (the origin), and all terms up to third order are retained:

$$\begin{aligned}\frac{dx_1}{d\tau} = &-[1+\beta_1(2+\beta_0)]\,x_1 - \beta_1\beta_3(1+\beta_0)\,x_2 \\ &+ \beta_1 x_1^2 + \frac{1}{2}\beta_1\beta_3(1+\beta_0)(\beta_3-2)x_2^2 \\ &+ \beta_1\beta_3(2+\beta_0)x_1x_2 - \beta_1\beta_3 x_1^2x_2 \\ &+ \frac{1}{2}\beta_1\beta_3(2+\beta_0)(2-\beta_3)x_1x_2^2 \\ &+ \frac{1}{6}\beta_1\beta_3(1+\beta_0)(-6+6\beta_3-\beta_3^2)x_2^3 + \ldots\ldots\end{aligned} \tag{11}$$

$$\begin{aligned}\frac{dx_2}{d\tau} = &\ \beta_5(2+\beta_0)x_1 - [\beta_4 - \beta_3\beta_5(1+\beta_0)]\,x_2 \\ &- \beta_5 x_1^2 - \frac{1}{2}\beta_3\beta_5(1+\beta_0)(\beta_3-2)x_2^2 \\ &- \beta_3\beta_5(2+\beta_0)x_1x_2 + \beta_3\beta_5 x_1^2x_2 \\ &- \frac{1}{2}\beta_3\beta_5(2+\beta_0)(2-\beta_3)x_1x_2^2 \\ &- \frac{1}{6}\beta_3\beta_5(1+\beta_0)(-6+6\beta_3-\beta_3^2)x_2^3 + \ldots\end{aligned} \tag{12}$$

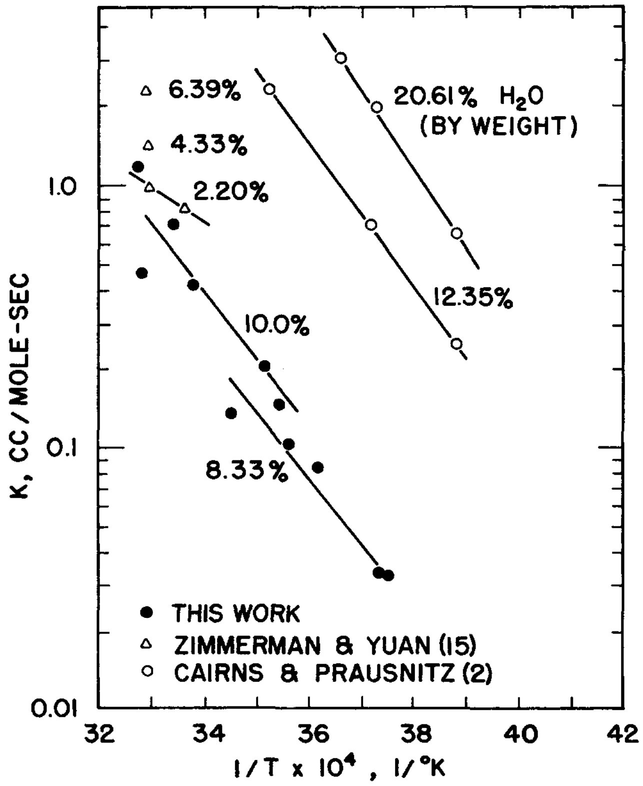

Fig. 3. Reaction rate constants.

This expansion allows us to approximate the complicated nonlinear functions by simpler third-order polynomials. However, in some cases the approximation changes the nature of the equations and therefore is invalid. For example, it is a simple matter to find sets of system parameters such that there is only one unstable steady state solution of the original nonlinear state equations, Equations (7) and (8), but more than one steady state solution of the approximate equations, Equations (11) and (12). Since some of these spurious solutions often are stable, the approximate equations can have different topological properties which will not correspond to the real system. This difficulty often can be avoided by neglecting the terms in x_2^3. Although this modification of the polynomial approximation is somewhat arbitrary, it usually leads to valid results.

In order to apply the analytical solution described in a previous paper (6), we must have a pair of complex conjugate roots with positive real parts, $\lambda = \alpha_0 \pm j\omega_0$. From the linearized equations, it can be shown that

$$\alpha_0 = -\frac{1}{2}\,[1+\beta_1(2+\beta_0) + \beta_4 - \beta_3\beta_5\,(1+\beta_0)]$$

and

$$\omega_0 = \left\{\beta_4\,[1+\beta_1(2+\beta_0)] - \beta_3\beta_5(1+\beta_0) - \frac{1}{4}[\,1+\beta_1(2+\beta_0) + \beta_4 - \beta_3\beta_5\,(1+\beta_0)]^2\right\}^{1/2} \tag{14}$$

The transformation matrix **P** is given in the appendix, and using this result we find that the approximate solution for the hydrolysis reaction is

$$x_1 = x_{1\mathrm{av}} + 2\,[\alpha_0 + 1 + \beta_1\,(2+\beta_0)]A_0 \cos\omega\tau + 2\omega_0\,A_0 \sin\omega\tau + \ldots \tag{15}$$

$$x_2 = x_{2\mathrm{av}} - 2\beta_5\,(2+\beta_0)\,A_0 \cos\omega\tau + \ldots. \tag{16}$$

The amplitude of the oscillation A_0 in these equations is given by

$$A_0^2 = \frac{+\,4\alpha_0}{K_1 + K_2 + K_3} \tag{17}$$

where K_1, and K_2, and K_3 are constants involving the system parameters and are given in the appendix. The frequency of the oscillation is

$$\omega = \omega_0 + \omega_1 \tag{18}$$

where

TABLE 1. SYSTEMS PARAMETERS

$A_f = 4.21 \times 10^{-3}$ mole/cc.(acetyl chloride), $B_f = 5.06 \times 10^{-3}$ mole/cc.(water), $V = 1{,}000$ cc., $q = 27.5$ cc./min. acetyl chloride + 64.0 cc./min. acetone and water, $k_0 = 3.37 \times 10^8$ (g. mole)$^{-1}$ (sec.)$^{-1}$ (cc.), $E = 12{,}000$ cal./mole, $(-\Delta H) = 22{,}400$ cal./mole, $C_P = 0.812$ (cal.)/(g.)(cc.), $\rho = 0.583$ g./cc., $T_f = 284$°K. $UA_c = 3.8$ (cal.)/(g.)(°C.), $C_{Pc}\,\rho_c = 0.887$ (cal.)/(cc.)(°C.), $q_c = 200$ cc./min., $T_c = 270$°K.

Steady State Results

$T_s = 295$°K., $A_s = 2.185 \times 10^{-3}$ mole/cc.

Dimensionless Parameters

$\beta_1 = 0.6676$, $\beta_3 = 20.4$, $\beta_4 = 4.31$, $\beta_5 = 0.2443$, $\alpha = 0.2$

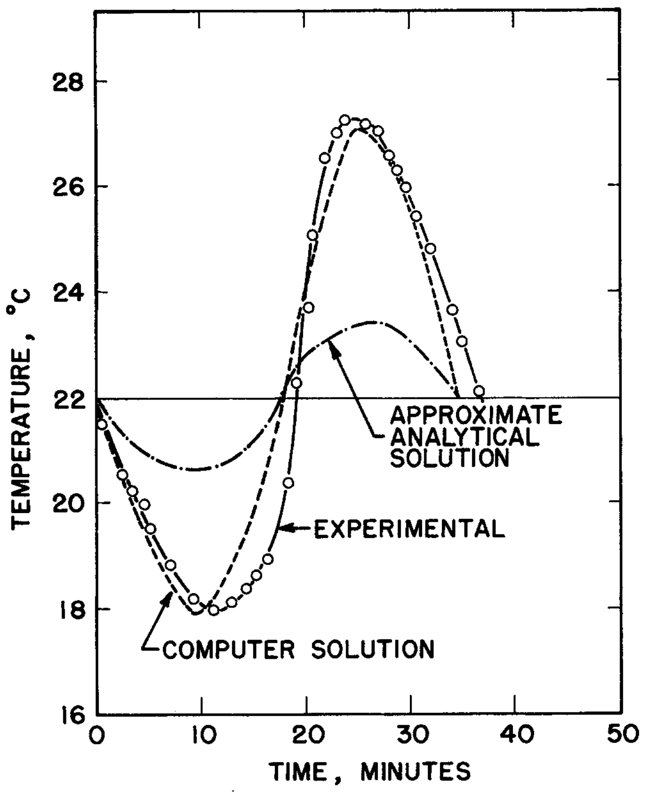

Fig. 4. Oscillating reactor temperature.

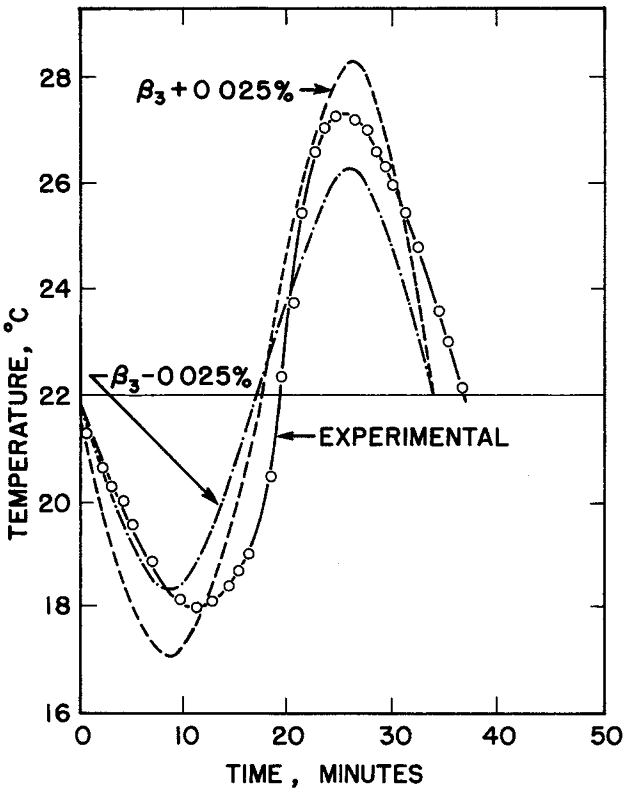

Fig. 5. Effect of parameter β_3 on the oscillations.

$$\omega_1 = \frac{A_0^2}{4}(K_4 + K_5 + K_6) \tag{19}$$

and K_4, K_5, and K_6 are given in the appendix. Also

$$x_{1av} = \frac{-2A_0^2}{\omega}\left[\alpha_0 + 1 + \beta_1(2 + \beta_0)\cdot (K_7 + K_8) - \frac{2A_0^2\,\omega_0}{\omega}K_9\right] \tag{20}$$

and

$$x_{2av} = \frac{2\beta_5(2 + \beta_0)A_0^2}{\omega}(K_7 + K_8)$$

where K_7, K_8, and K_9 are listed in the appendix.

EXPERIMENTAL EQUIPMENT

Preliminary calculations based on the solution above indicated that it would be difficult to design a laboratory reactor which would generate oscillations under normal conditions. These estimates showed that it would be necessary both to operate at higher feed concentrations than those studied by Cairns and Prausnitz and to cool the reactor below ambient conditions. Hence, an attempt was made to design a flexible reactor system which could be used to investigate oscillatory behavior.

A schematic diagram of the equipment is shown in Figure 2. The reactor was a 2-liter polyester vessel which was resistant to the highly corrosive reactant and product materials. A Laboratory Lightning Mixer with a polyester coated impeller supplied sufficient agitation. The mixer speed was controlled by using a Variac, and the submerged cooling coils acted as baffles.

Acetone and water were mixed to the desired proportions in a 7-gal. tank and eventually forced into the reactor under nitrogen pressure. Flow rates of 50 to 600 cc./min. were achieved in the ¼-in. copper tubing feed line, and these were controlled with a stainless steel needle valve. Provisions were made so that part of the copper feed line could be immersed in the cooling bath. Approximately 24 cc./min. of acetyl chloride were supplied to the reactor by using a Pyrex constant-head delivery funnel having a 1-liter reservoir.

A mixture containing 10% methanol, 25% ethylene glycol, and 65% water by volume was used as the coolant material. This ternary mixture has a low viscosity and a high thermal capacity in the temperature range of interest. Dry ice was added directly to about 5 liters of coolant material contained in a reservoir. A 1/16 hp. epoxy encased March submersion pump forced the cold fluid through a 17 ft. long coil of ¼-in. diameter 316 stainless steel tubing located in the reactor. It was normally possible to maintain the coolant supply temperature to within 1°F. with this cooling system.

The reactor temperature was measured both with a mercury thermometer having a range of −2° to 100°C. graduations and with a YP1 thermistor unit having a range of −50° to 350°F. and accurate to within 1°F. The thermistor was placed in a small 316 stainless steel well, and the overall time constant of this assembly was approximately 3.5 sec. The dynamics of both measuring systems had no effect on the low frequency oscillations generated by the system. No attempt was made to measure the reactor concentration since the conductivity method described by Cairns and Prausnitz cannot be easily extended to concentrated mixtures (*2, 11, 13*).

EXPERIMENTAL MEASUREMENTS

Since the concentrations of acetyl chloride used in the present investigation were much higher (3 to 4N) than those used in the previous kinetic (10^{-5} to 10^{-2}N) or dynamic studies (0.5N), and since the results of Cairns and Prausnitz (*2*) showed that the reaction rate was highly dependent upon the dielectric constant of the reacting medium, it was necessary to obtain new estimates of the kinetic parameters. Feed mixtures of 8.33 and 10.0 wt. % water in acetone and an α ratio of 0.2 were introduced into the reactor, and the reactor temperature was measured as a function of time. Once steady state conditions had been obtained, it was possible to calculate a value of the rate constant k from Equation (5) after we substituted the measured value of the final steady state temperature and the other known inputs for the system. Then ln k was plotted against the reciprocal of the absolute value of the reactor temperature, and Equation (4) was used to find the

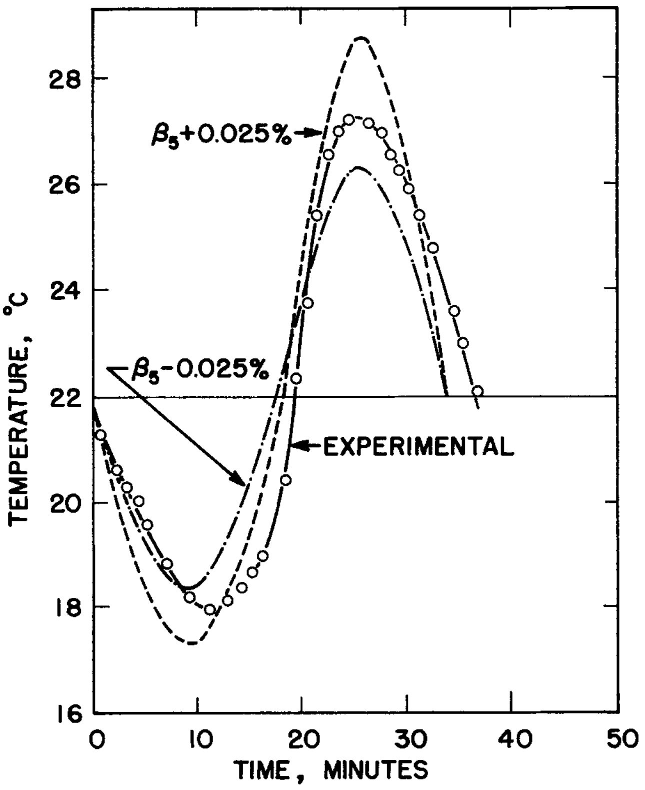

Fig. 6. Effect of parameter β_5 on the oscillations.

values of the frequency factor k_o and the activation energy E. The experimental results are plotted in Figure 3. Although the concentration of water is used to identify the curves, the amount of solvent also varies between the different runs. The measured reaction rate constants for these concentrated mixtures are about an order of magnitude lower than those published previously for dilute solutions because of the difference in the dielectric constants of the reacting mixtures.

After the kinetic parameters had been established and the value of UA_c measured for a case where there was no reaction, it was possible to change some of the operating variables so that the oscillator criteria were satisfied. The pseudo steady state portion of the measured temperature response for one of the runs is plotted in Figure 4. Different initial conditions gave the same oscillatory output. The numerical solutions of the nonlinear system equations and the approximate analytical solutions for this run, corresponding to the parameters listed in Table 1, also are plotted on this graph. For these parameters, the difference between the time average outputs and the steady state values are negligible; that is, $x_{1\,av} \simeq 4.4 \times 10^{-3}$.

DISCUSSION OF RESULTS

The agreement between the numerical solutions of the CSTR equations and the experimental data can not be expected to be perfect because of experimental errors, the assumptions inherent in the dynamic model, and an imprecise knowledge of some of the system parameters. Rather than obtaining better estimates of these quantities or refining the experimental apparatus, we decided to study the sensitivity of the numerical solutions to small changes in the system constants. The results for 0.025% variations in β_3 and β_5 are presented in Figures 5 and 6. These curves bracket the data fairly well and indicate that it probably would be a very difficult matter to improve the fit very much.

The approximate analytical solutions show a much poorer agreement with the experimental values than the numerical solutions because the cubic terms in the temperature deviations were neglected in the Taylor series expansions in Equations (11) and (12). A more complete explanation of the discrepancy caused by this approximation will be presented elsewhere (3). However, the results show that the analytical solutions provide at least qualitative predictions of the oscillating outputs, and therefore that it should be possible to obtain useful information from studies on this model. In particular, they can be used to decide how to modify the design of the system to obtain significant shifts in the time average conversion. Also, they provide some justification for extending the theoretical studies on chemical oscillators to complex reactions and other cases where significant improvements in the economic performance might be obtained.

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NOTATION

A, A_f = composition of acetyl chloride in the reactor and feed, respectively
A_c = area of cooling coil
A_0 = amplitude of the oscillation
B, B_f = composition of water in the reactor and feed, respectively
C_p = heat capacity
E = activation energy
$(-\Delta H)$ = heat of reaction
K = constant defined by Equation (4)
K_1 to K_9 = constants in analytical solution, see appendix
k = reaction rate constant
k_0 = frequency factor
$\mathbf{P}$ = transformation matrix, see appendix
q = flow rate
R = gas constant
T = temperature
t = time
U = overall heat transfer coefficient
V = reactor volume
x_1 = dimensionless composition, see Equation (6)
x_2 = dimensionless temperature, see Equation (6)

Greek Letters

α = constant defined by Equation (6)
α_0 = real part of characteristic root, see Equation (13)
β_0, β_1, β_3, β_4, β_5 = constants defined by Equation (6)
λ = characteristic roots
ρ = density
τ = dimensionless time, see Equation (6)
ω = frequency of nonlinear oscillator
ω_0 = imaginary part of characteristic root, frequency of linear oscillator
ω_1 = frequency correction factor in analytical solution

Subscripts

av = average
c = coolant
f = feed
s = steady state

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APPENDIX: TERMS IN ANALYTICAL SOLUTION

Transformation Matrix

$$\mathbf{P} = \begin{pmatrix} P_{11} & P_{12} \\ P_{21} & P_{22} \end{pmatrix} = \begin{pmatrix} 2\,\alpha_0 + 2[1 + \beta_1(2 + \beta_0)] & 2\omega_0 \\ -\,2\beta_5(2 + \beta_0) & 0 \end{pmatrix}$$

Constants in Solution

$$\begin{aligned} K_1 = &-\,2\beta_3\beta_5\,\{3[1 + \alpha_0 + \beta_1(2 + \beta_0)]^2 + \omega_0{}^2\} \\ &-\,3\beta_3\beta_5{}^2(2 + \beta_0)^2(2 - \beta_3)[1 + \alpha_0 + \beta_1(2 + \beta_0)] \end{aligned}$$

$$\begin{aligned} K_2 = &-\,4\beta_1\beta_3\beta_5(2 + \beta_0)[1 + \alpha_0\beta_1(2 + \beta_0)] \\ &-\,\beta_1\beta_3\beta_5{}^2(2 + \beta_0)^3(2 - \beta_3) \end{aligned}$$

$$\begin{aligned} K_3 = &\,\beta_3\beta_5[1 + \alpha_0 + \beta_1(2 + \beta_0)] \\ &\{4[1 + \alpha_0 + \beta_1(2 + \beta_0)] + \beta_5(2 + \beta_0)^2(2 - \beta_3) \end{aligned}$$

$$\begin{aligned} K_4 = &-\,\beta_3\beta_5\omega_0\,\{4[1 + \alpha_0 + \beta_1(2 + \beta_0)] \\ &+\,\beta_5(2 + \beta_0)^2(2 - \beta_3)\} \end{aligned}$$

$$\begin{aligned} K_5 = &\,[2\beta_1\beta_3\beta_5(2 + \beta_0)/\omega_0]\,\{3[1 + \alpha_0 + \beta_1\,\cdot \\ &(2 + \beta_0)]^2 + \omega_0{}^2 + 3\beta_5(2 + \beta_0)^3\,\cdot \\ &(2 - \beta_3)[1 + \alpha_0 + \beta_1(2 + \beta_0)]\} \end{aligned}$$

$$\begin{aligned} K_6 = &-\,(\beta_3\beta_5/\omega_0)[1 + \alpha_0 + \beta_1(2 + \beta_0)] \\ &\{6[1 + \alpha_0 + \beta_1(2 + \beta_0)]^2 + \omega_0{}^2 \\ &+\,3\beta_5{}^2\beta_3(2 + \beta_0)^2[1 + \alpha_0 + \beta_1(2 + \beta_0)]\} \end{aligned}$$

$$\begin{aligned} K_7 = &\,(\beta_1/\omega_0)\,\{[1 + \alpha_0 + \beta_1(2 + \beta_0)]^2 + \omega_0{}^2\} \\ &+\,(\beta_1\beta_3\beta_5{}^2/2\omega_0)(1 + \beta_0)(2 + \beta_0)^2(\beta_3 - 2) \\ &-\,(\beta_1\beta_3\beta_5/\omega_0)(2 + \beta_0)^2[1 + \alpha_0 + \beta_1(2 + \beta_0)] \end{aligned}$$

$$\begin{aligned} K_8 = &\,\{[1 + \alpha_0 + \beta_1(2 + \beta_0)]/2\omega_0(2 + \beta_0)]\} \\ &\{2\beta_3\beta_5(2 + \beta_0)^2[1 + \alpha_0 + \beta_1(2 + \beta_0)] - 2\omega_0{}^2 \\ &-\,2[1 + \alpha_0 + \beta_1(2 + \beta_0)]^2 \\ &-\,\beta_3\beta_5{}^2(2 + \beta_0)^2(1 + \beta_0)(\beta_3 - 2)\} \end{aligned}$$

$$\begin{aligned} K_9 = &\,[1/2(2 + \beta_0)]\,\{2\beta_3\beta_5(2 + \beta_0)^2 \\ &[1 + \alpha_0 + \beta_1(2 + \beta_0)] - 2\omega_0{}^2 \\ &-\,2[1 + \alpha_0 + \beta_1(2 + \beta_0)]^2 \\ &-\,\beta_3\beta_5{}^2(2 + \beta_0)^2(1 + \beta_0)(\beta_3 - 2)\} \end{aligned}$$

Single Drop Breakup in Developing Turbulent Pipe Flow

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An experimental study was made on the breakup of a single drop in two phase liquid-liquid developing turbulent pipe flow. The predominant breakup mechanism was controlled by dynamic pressure forces, and the drop breakup was characterized by one or two smaller drops breaking off a larger drop.

The effect on the increase in interfacial area and the resulting size distribution of the fragmented drops of drop velocity, interfacial tension, initial drop size, and the distance down the pipe was studied. Regression analysis was used to determine the significant effects. Most of these regression equations had significant cubic and two-factor interactions, which is indicative of the complexity of drop breakup.

The size distribution of the fragmented drops can be approximated by a normal distribution. However, the steady state distribution was not fully developed in the work here.

In many two phase liquid-liquid operations in chemical engineering, where mass is being transferred between phases, the mass transfer step is the rate determining step in the operation. The mass transfer of species A depends upon the mass transfer coefficient, the interfacial area, and the chemical potential of A in each of the two phases,

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