

TRANSIENT RESPONSE OF AN IMPERFECTLY MIXED  
CONTINUOUS FLOW STIRRED TANK REACTOR  
TO SIMULTANEOUS STEP CHANGES  
IN INLET CONCENTRATION AND FLOW RATE

by

Hoagy Kim

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A thesis submitted to the Faculty and the Board of Trustees of the Colorado School of Mines in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Chemical and Petroleum-Refining Engineering.

Signed: Hoagy Kim  
Hoagy Kim

Golden, Colorado

Date: Oct 14, 1970

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Approved: Philip F. Dickson  
Dr. Philip F. Dickson

James H. Gary  
Dr. James H. Gary  
Head of Department

Golden, Colorado

Date: Oct 14, 1970

To the memory of my beloved brother

Yonggi Kim, M.D.

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ABSTRACT

Nonideal behavior of an imperfectly mixed continuous flow tank reactor under isothermal condition was investigated by observing the transient response of the outlet concentration to simultaneous step changes in inlet flow rates and concentrations. The saponification reaction of ethyl acetate with sodium hydroxide was adopted for the experimental work.

Residence time distribution at various mixing levels was obtained from tracer information, and mixed models of perfectly stirred tank and plug flow reactors in series were adopted for macromixing models. It was shown that the conversion decreased with an increase of the mixing rate.

The perturbation method and the method of characteristics were found useful in obtaining the solutions for the transient concentrations.

Comparing the experimental transient concentration with those of a plug flow reactor, a perfectly stirred tank reactor, and the mixed model show deviation from perfect mixing and confirms the existence of late mixing.

The mixing pattern is a strong function of the reactor

geometry. The theory developed herein can be applied to other reactor geometries by obtaining the tracer response of the system in question. The model then allows prediction of the reaction conversion for this new system.

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## INTRODUCTION

All flow reactors deviate to some extent from the ideal cases of perfect mixing and plug flow. In a perfectly stirred tank reactor, the composition of the reacting fluid would have a uniform value throughout the reactor and equal to the output composition. Perfect stirring can be achieved at a very high stirring rate. However, the power cost for the stirring may be an important economic factor, and the assumption of perfect mixing cannot be satisfied in many cases.

One of the earliest works on the effect of mixing on the reactor performance was presented by Bodenstein and Wolgast<sup>(1)</sup>, who studied the difference between perfect mixing and plug flow. In 1935, MacMullin and Weber<sup>(2)</sup> presented a pioneering work, in which they studied the residence time distribution of a system of tanks in series with short circuiting. Methods of modeling of the flow system were first presented by Danckwerts<sup>(3)</sup>, who defined certain distribution functions, such as external and internal age distributions, which give information on the mixing characteristics in vessels. The methods of experimental measurement of age-distribution functions and their interpretation to detect dead regions, by-

passing, and non-uniform regions are lucidly summarized by Levenspiel<sup>(4)</sup>, Himmelblau and Bischoff<sup>(5)</sup>, Levenspiel and Bischoff<sup>(6)</sup>, and others.

In order to study the behavior of the actual reactors, two modes of mixing, namely micromixing and macromixing, should be considered. In micromixing individual molecules are free to move about and intermix, while with macromixing molecules are visualized as grouped together in aggregate or packets.

Many of the main features of macromixing effects can be represented by single coefficient dispersion models. Levenspiel and Bischoff<sup>(7)</sup> presented dispersion models for second-order reaction and Fan, and Bailie<sup>(8)</sup> presented those for the reaction orders of  $\frac{1}{4}$ ,  $\frac{1}{2}$ , 2, 3, as well as complete sets of concentration profiles through the reactor section.

Degree of segregation as a measure of micromixing was defined by Danckwerts<sup>(9)</sup>. The two limits of micromixing, namely complete segregation and maximum mixedness, were defined by Danckwerts<sup>(9)</sup> and Zwietering<sup>(10)</sup>, respectively.

Reactor performance can be predicted effectively by mixed models which consist of interconnected flow regions with various modes of flow such as plug flow regions, perfectly stirred regions, dead space, and crossflow. Models are frequently determined with the help of age-distribution functions obtained from the tracer runs on real reactors, and these models are then used to predict the behavior of

the real reacting systems.

Age-distribution functions obtained from the tracer studies give information on macromixing but not on micromixing. Therefore, tracer curves can predict the performance of the reactors for the linear processes (first-order reaction) where interactions between fluid elements do not affect the reaction rate.

For the nonlinear processes, the tracer curves give only the limits on conversion; and information on micromixing as well as macromixing is necessary for the complete description of the reactor behavior. Complete segregation and maximum mixedness are two limits for micromixing. The former considers each fluid element to be totally isolated from the other elements, while the latter considers the elements to mix at the earliest possible moment. Determining the degree of segregation will give a semiquantitative idea of the micromixing behavior in a vessel, but it cannot be related qualitatively to the performance of a reactor.

The first work to describe the behavior of a real stirred tank reactor for a linear process under isothermal conditions with a mixed model was presented by Cholette and Cloutier<sup>(11)</sup>. From the result of tracer runs, they found that their reactor was best fitted by a model consisting of a backmix region and of a dead space with a portion of the fluid bypassing the vessel. They also presented these parameters as functions of the mixing rate. Bartock et al<sup>(12)</sup>

also presented work with linear systems. For a second-order reaction, experimental work has been presented by LaRosa and Manning<sup>(13)</sup>, who found that measured rates of reaction decreases with the increase of degree of mixing and correlated the degree of segregation with rpm. A number of further works have been presented.<sup>(14,15,16,17)</sup>

Lelli<sup>(18)</sup> suggested that the residence time distribution obtained from a reacting-tracer isothermal first-order system with variable inlet stream concentrations may more accurately simulate actual systems of interest. Mathematical theory on the residence time distribution for reacting systems was first considered by Nauman.<sup>(19)</sup>

Other investigators made an attempt to describe the actual reactor with stochastic models rather than deterministic ones. Krambeck et al<sup>(20)</sup> presented a mathematical theory which considers the probabilities of flows flowing within n-stirred tanks arbitrarily connected by interstage flow. It seems that numerous stochastic variables make this method quite impractical.

In this work, the nonideal behavior of an imperfectly mixed reactor was investigated by observing the transient response of the outlet concentration to step upsets in inlet flow rates and concentrations for a second-order reaction under isothermal conditions.

First, tracer runs were performed to find age-distribution functions in order to predict the mixed model. Because the reaction was nonlinear, two extreme models

were taken to bound the conversion for macromixing. The other limit for complete segregation with the known residence time distribution was calculated. Mathematical solutions for the transient concentration for a perfectly stirred tank, a plug flow reactor, and a mixed model were compared with the experimental data.

## EXPERIMENTAL WORK

Figure 1 shows the schematic diagram for the experimental system. The reagents were stored in 5-gallon bottles and fed, through a constant temperature bath at 80°F., to the top of the reactor by air controlled with a pressure regulator. Flow rates were measured by Fischer & Porter rotameters ranging in capacity from 20 to 155 cc per minute. The flow lines were  $\frac{1}{4}$ -inch Polyflo tubing.

The geometry of the reactor is shown in Figure 2. In order to set the pH probe in an upright position for maximum efficiency, the reactor top was sealed, and an overflow reactor was adopted. The reactor consisted of a 1225 cc stainless-steel cylindrical vessel which was 4 inches in diameter and 5  $\frac{3}{4}$  inches in height, mounted with four baffles ( $\frac{13}{32}$ -inch wide and 4-inch high). The impeller was a three-blade stirrer 1 inch in diameter and was mounted 2 inches from the bottom of the reactor. The saponification reaction of ethyl acetate with sodium hydroxide was adopted for the experimental work.

Following are the procedures taken for tracer runs and transient reaction runs.

Figure 1. Schematic Flow Diagram

1. Air
2. Pressure Regulator
3. Pressure Gage
4. Feed Bottles
5. Three-Way Valves
6. Rotameters
7. Constant Temperature Bath
8. Flow Cell with pH Electrode
9. Variable Speed Motor and stirrer
10. Tachometer
11. Speed Control
12. Sample
13. pH Meter
14. Recorder
15. Reactor System

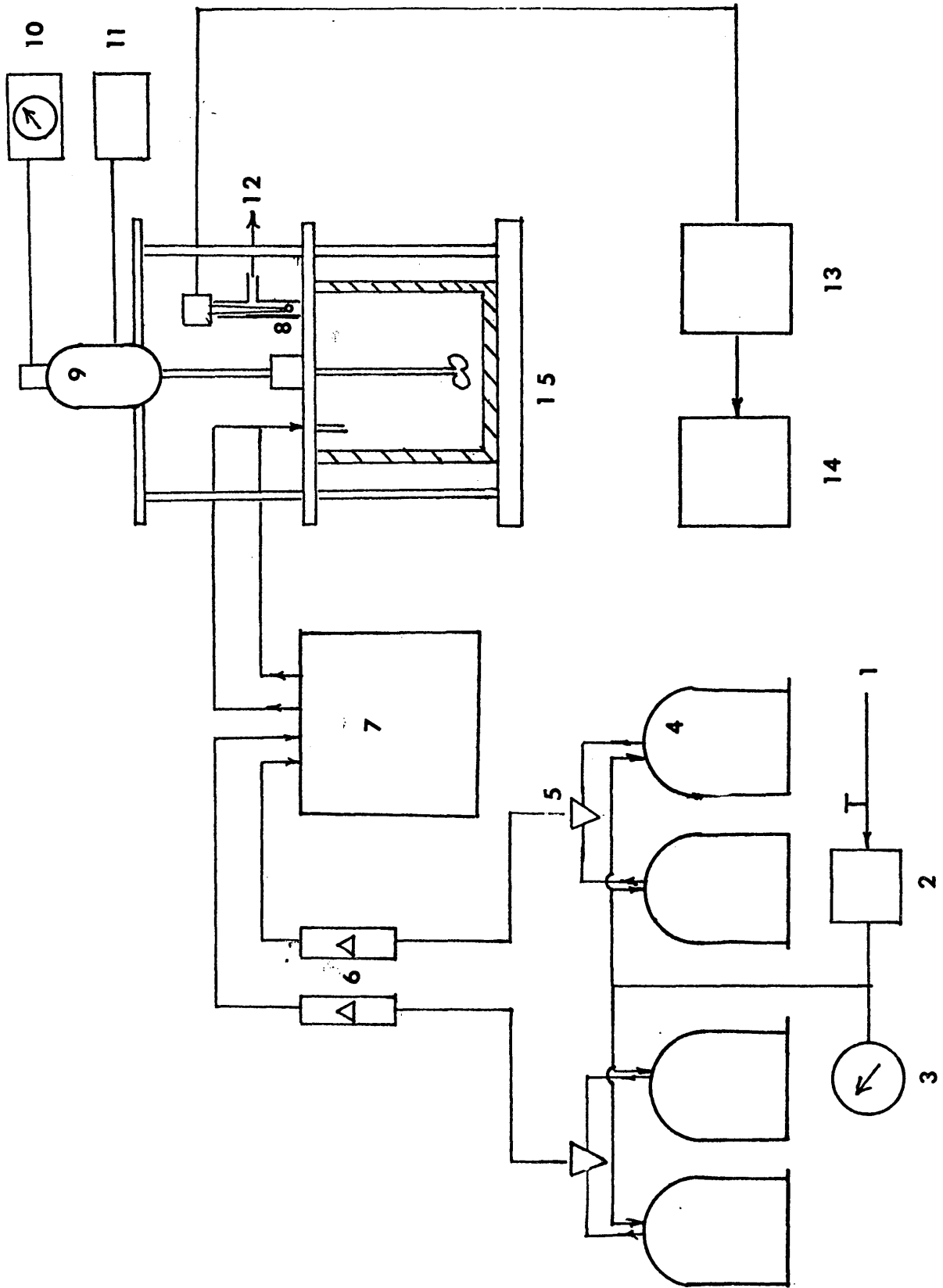
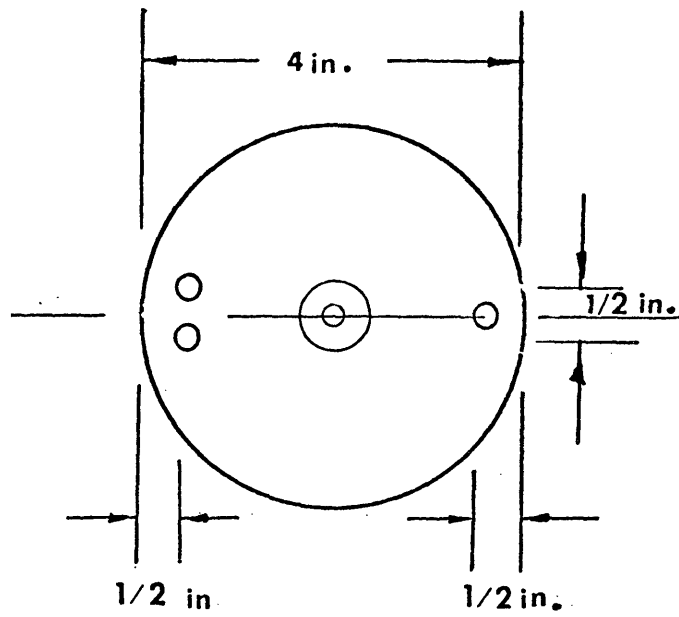
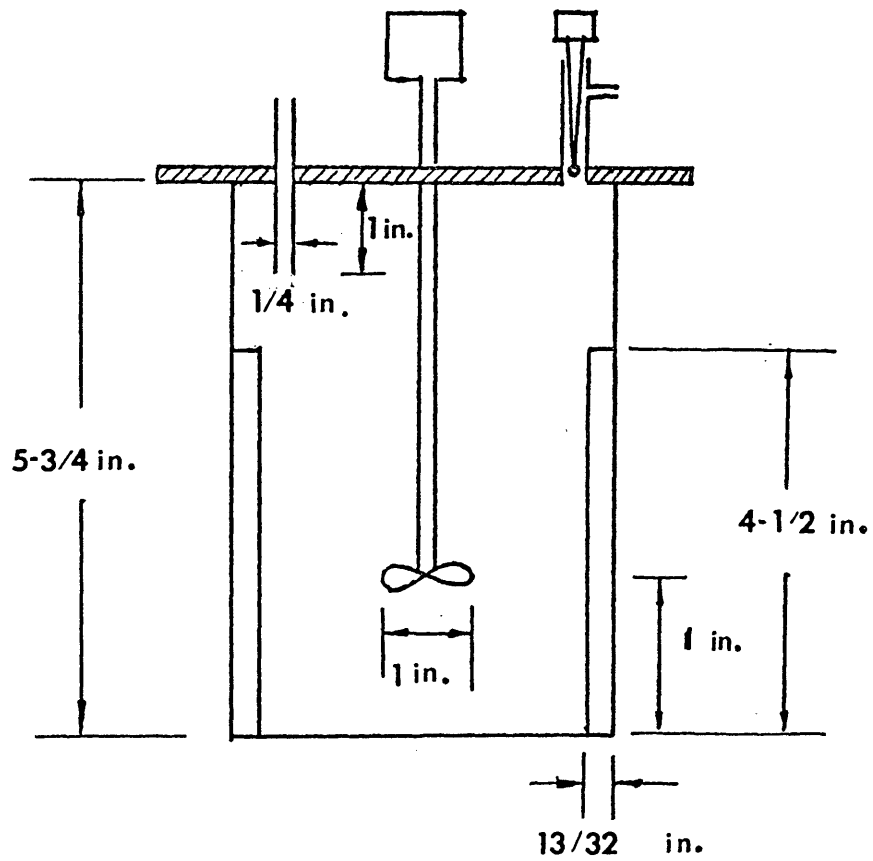


Figure 2. Reactor Geometry



TOP VIEW



SIDE VIEW

### Tracer Runs

First, distilled water from two of the storage bottles were introduced to the empty reactor through the two reactant tubes at 80 ml per minute each. When the reactor was filled, tracer was introduced by turning the three-way valves to the other two bottles containing 0.1 N sodium hydroxide. The resulting transient sodium hydroxide concentration at the outlet was obtained on a Mosley 7100B strip-chart recorder.

### Transient Reaction Runs

Solutions of 0.1 N and 0.05 N sodium hydroxide were prepared and stored in two different bottles connected to one of the inlet lines, and 0.1 N ethyl acetate solution was stored in the other two bottles. Initially, 0.1 N sodium hydroxide and 0.1 N ethyl acetate were passed through the reactor at the flow rate of 80 ml per minute each. When the steady state was observed in the recorder, a sample was withdrawn to known amount of excess 0.1 N HCl solution. After collecting the sample, sodium hydroxide concentration was upset to 0.05 N by using the three-way valve, and ethyl acetate flow rate was raised to 100 ml per minute. After the new steady state was reached, a sample was collected to known amount of excess 0.1 HCl. Each sample was titrated with 0.01 N sodium hydroxide solution immediately after being withdrawn from the outlet of the reactor, and the tran-

sient concentration of sodium hydroxide was obtained by the 7100B strip-chart recorder. At the concentration employed in this work, heat generation was negligible and the system was isothermal.

TRACER INFORMATION

For the step input of tracer with the concentration  $C_0$ , the response of the outlet concentration for the two ideal cases of perfectly mixed continuous stirred tank (CSTR) and plug flow (PF) are shown in Figure 3.

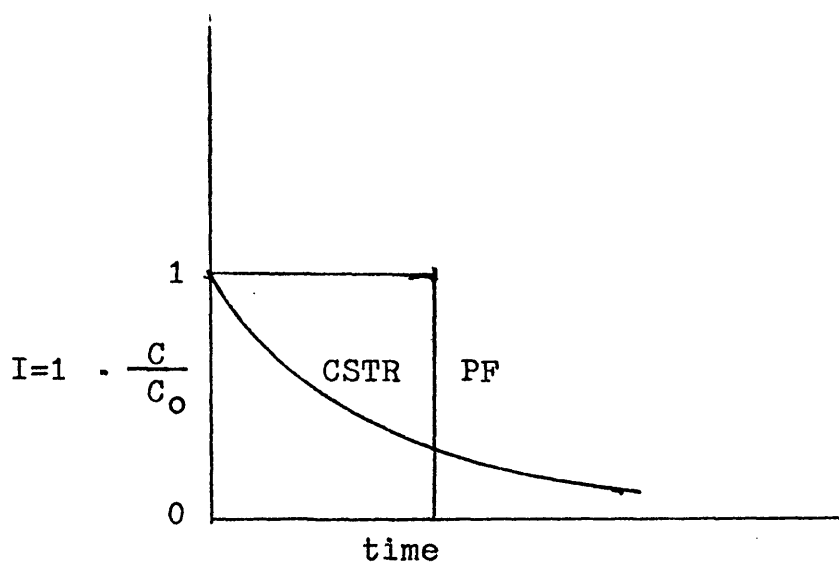


Figure 3. Response at the outlet to input tracer for CSTR and PF

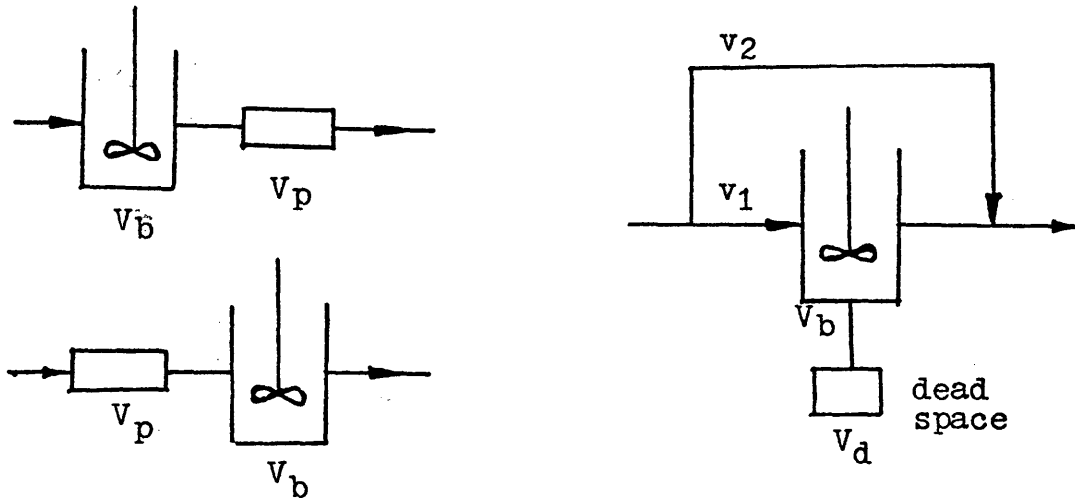
Actual responses of an imperfectly mixed stirred tank reactors would lie somewhere in between these two limits. In this work  $\underline{I}$  curves as shown in Figure 3 at the various mixing level were obtained by the continuous injection of sodium hydroxide tracer (0.1 N) to the inflow line (flow rate of 0.16 cc per minute, initially water). These curves were compared with the responses of known mixed models, some of which are shown in Figure 4.

The experimental data are shown in Appendix (p.60-62) and the residence time distributions ( $\underline{I}$  curves) are shown in Figures 5a through 5d. The apparent delay action during the initial portion and subsequent exponential change in output response to the tracer injection resulted in the choice of the model of PF and CSTR in series as shown in Figure 4a. However, whether PF portion precedes CSTR or vice versa could not be determined from the tracer information alone, as mentioned in the introduction.

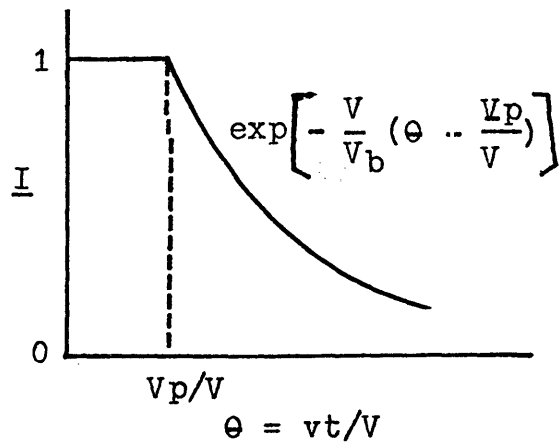
It was shown from the experiment that the fraction of the plug flow portion ( $V_p/V$ ) consistently decreased with the increase of the stirring rate (rpm). Figure 6 shows this trend.

It appeared that  $\log(V_p/V)$  was a linear function of rpm, and the following correlation was drawn by using the least square techniques:

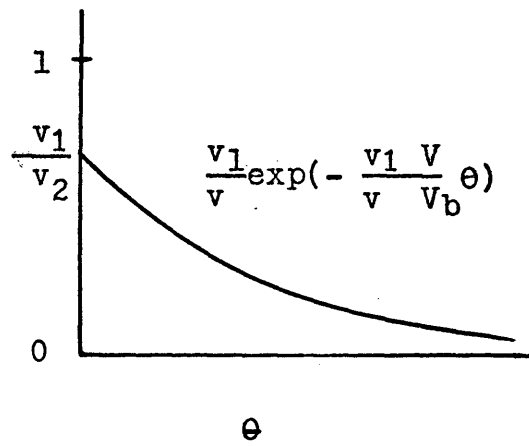
$$\log (V_p/V) = -1.075 \times 10^{-3}(\text{rpm}) + 2.001 \quad (1)$$



a1. S-T Model

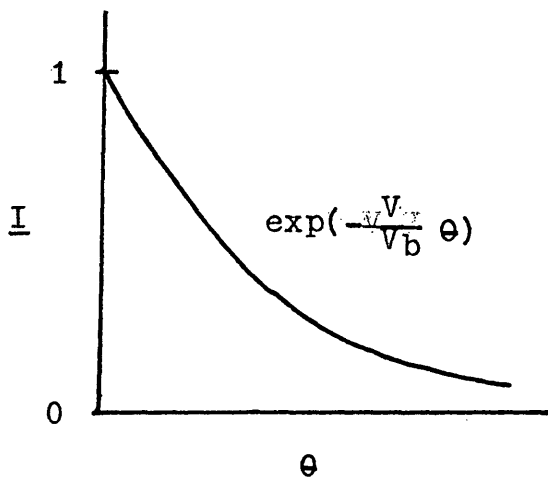
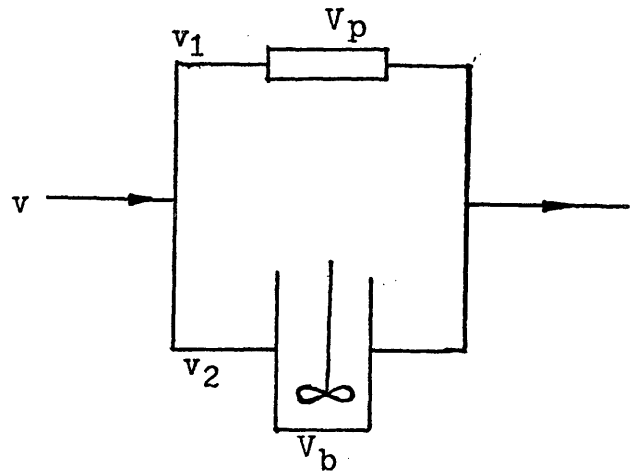
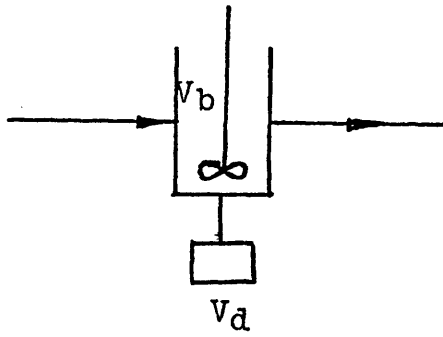


a2. T-S Model

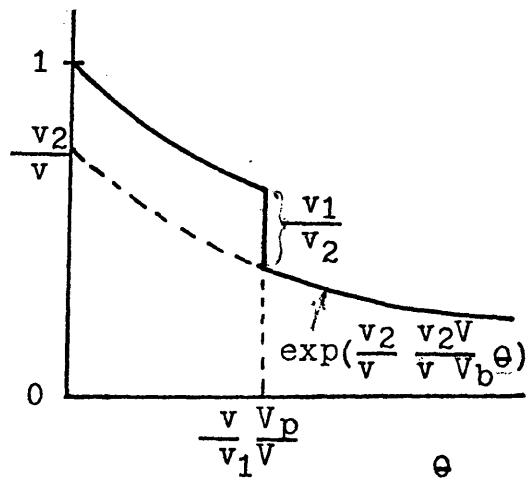


b. Cholette & Cloutier Model

Figure 4. Some simple mixed models and their residence time distribution functions: in part from Levenspiel (4)



c. CSTR with dead space



d. PF and CSTR in parallel

Figure 4 (continued)

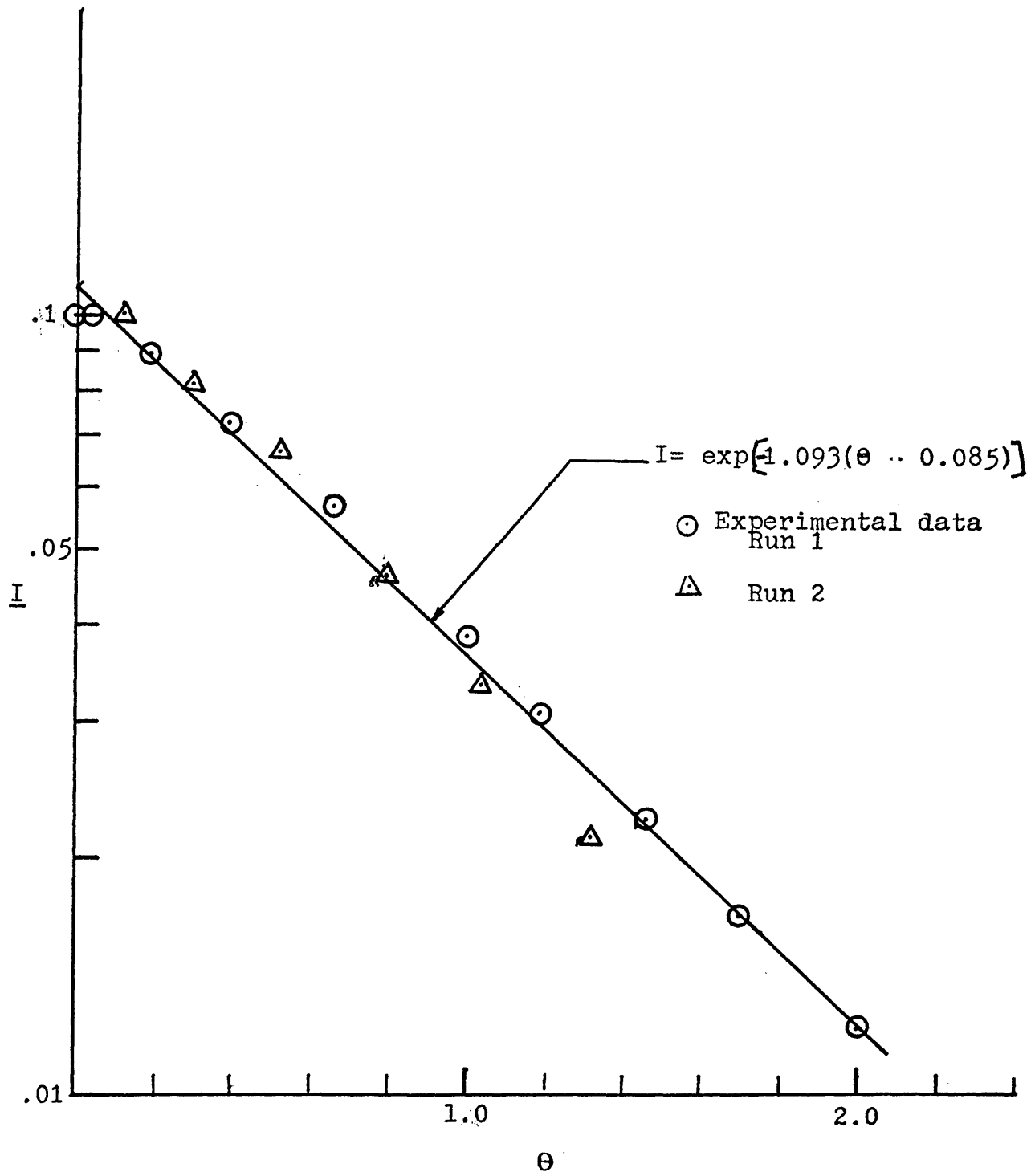


Figure 5a. Residence time distribution function  $I$  at no mixing

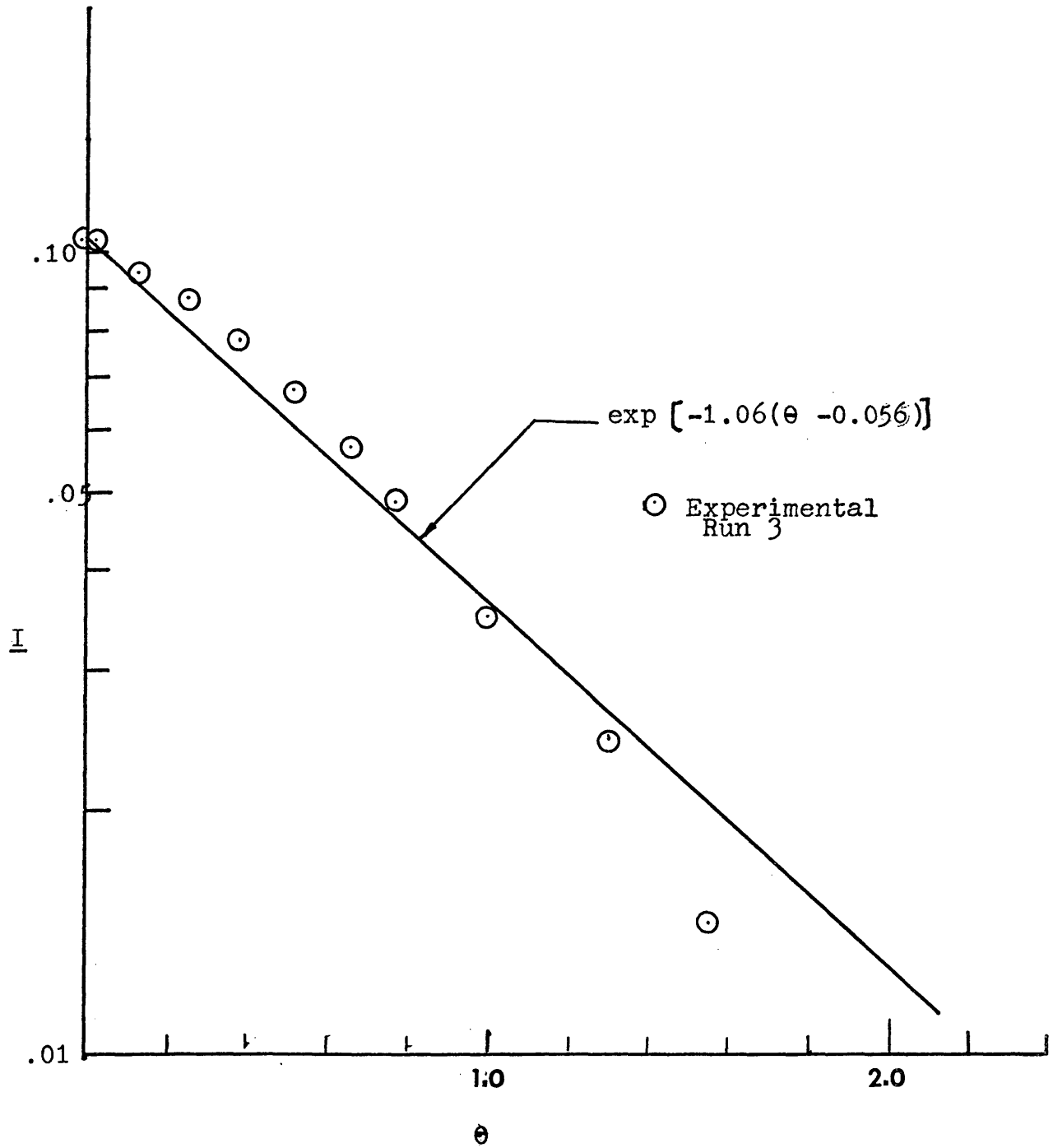


Figure 5b. Residence time distribution function  $I$   
at 194 rpm

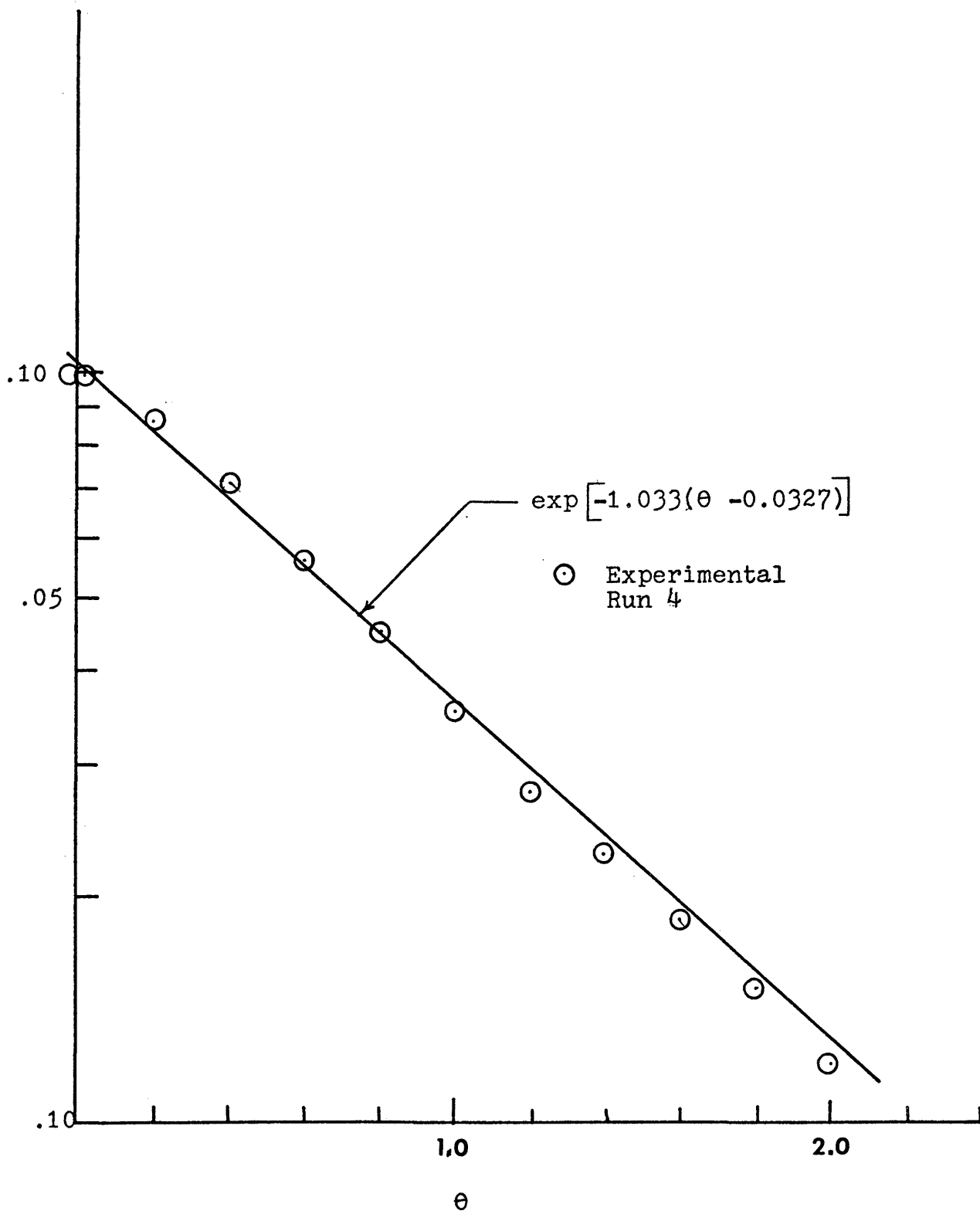


Figure 5c. Residence time distribution function  $I$   
at 279 rpm

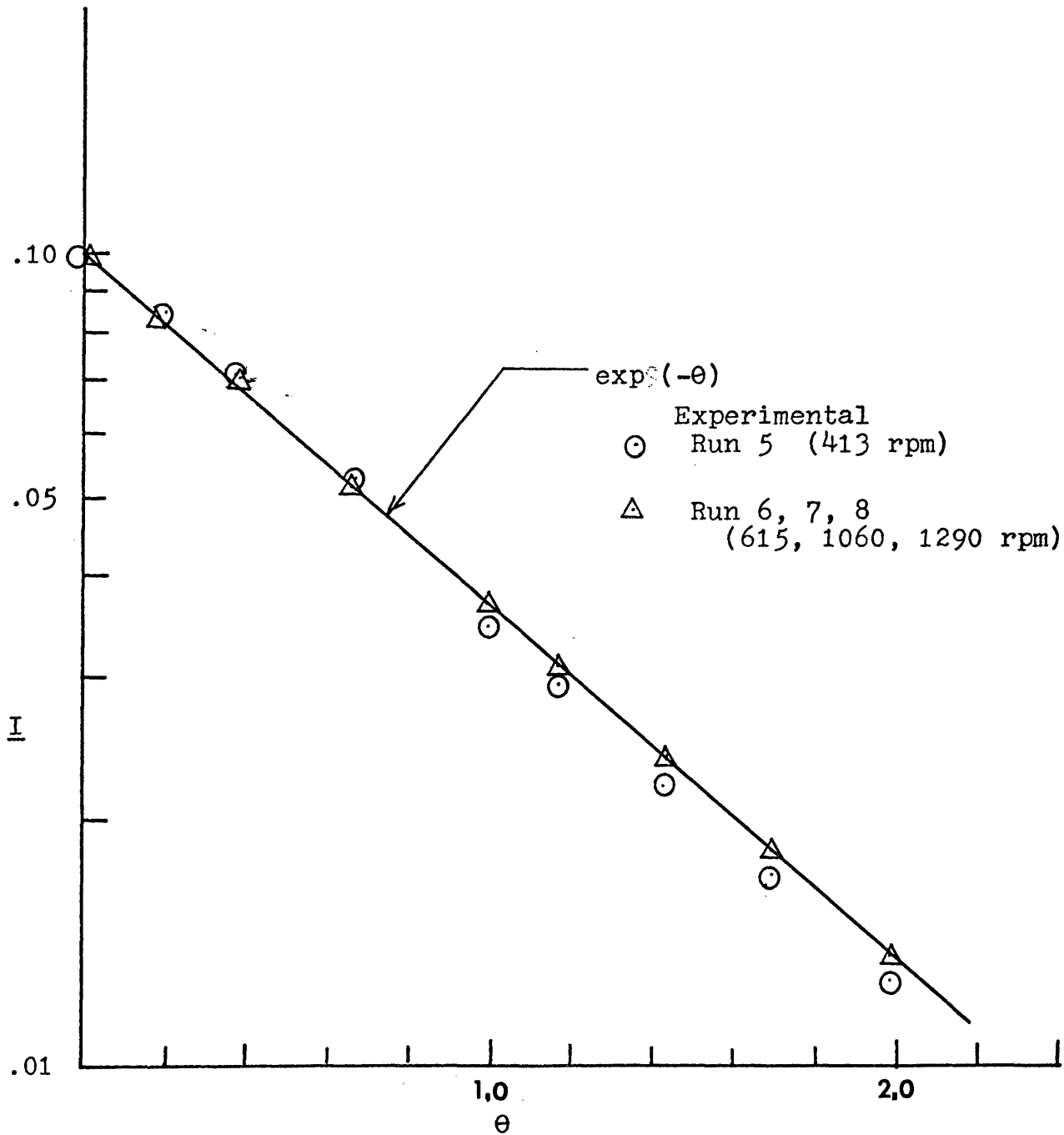
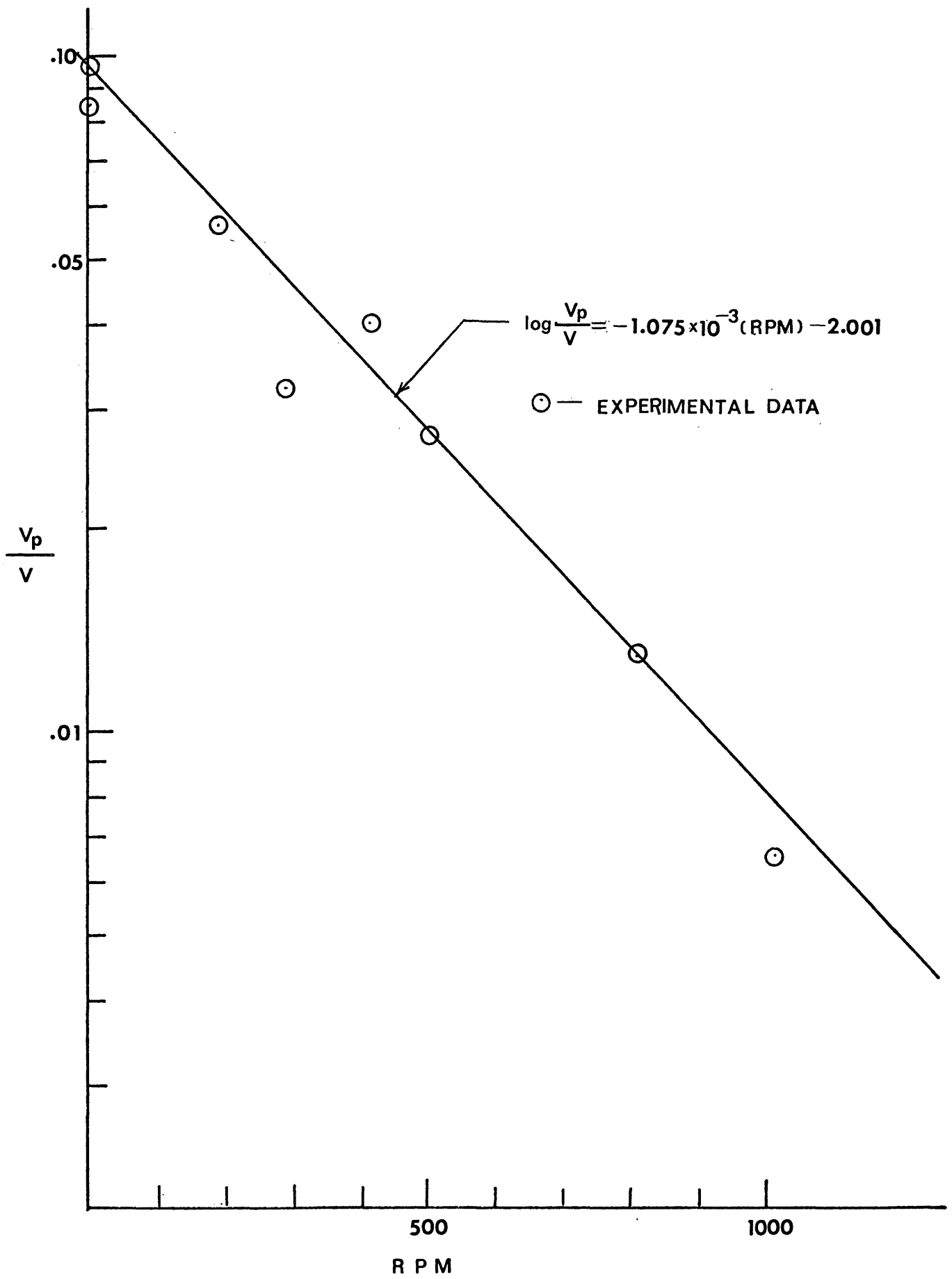


Figure 5d. Residence time distribution function  $I$   
over 413 rpm

Figure 6. Fraction of the plug flow portion  
as a function of mixing rate



The exponential change of response to the tracer as shown in the equation in Figure 4a was in excellent agreement with that obtained from the experiment for rpm range over 279. For the lower rpm some discrepancies were shown. However, the apparent delay action coupled with the trend of decreasing  $V_p/V$  with the rpm justify the same model for this region also.

There was negligible error in NaOH concentration and approximately  $\pm 3\%$  fluctuations in flow rates. The error in time delay (approximately 5 seconds) from the point where the tracer was introduced to the point where it was detected was obtained by the responses at high rpm where perfect mixing is reached. Over 615 rpm the experimental responses essentially did not change and the delay in these responses was considered the error, and this amount was subtracted in each run.

LIMITS ON CONVERSION

In the previous section, it was found that the reactor system adopted in this study closely followed the arrangement of perfectly stirred tank--plug flow (S-T) or plug flow--perfectly stirred tank (T-S) model as shown in Figure 4a.

These models give conversions for macromixing. For a first-order reaction ( $A \longrightarrow$  products,  $dC_A/dt = -kC_A$ ), both models will give same conversion by material balance.

$$X_A = 1 - \frac{C_{A^e}}{C_{A_0}} = 1 - \frac{e^{-k\bar{\tau}_p}}{1 + k\bar{\tau}_p} \quad (2)$$

For a second-order reaction ( $A + B \longrightarrow$  products,  $dC_A/dt = -kC_A C_B$ ), material balances give different conversions for T-S and S-T models:

For S-T model,

$$X_A = 1 + \frac{1 - \sqrt{4C_{A_0}k\bar{\tau}_p + 1}}{2C_{A_0}k\bar{\tau}_p + C_{A_0}k\bar{\tau}_p (-1 + \sqrt{1 + 4C_{A_0}k\bar{\tau}_p})}$$

if  $C_A = C_B$  (3)

$$X_A = 1 - \frac{N}{C_{A0} \left( 1 + \frac{N - C_{Ai}}{C_{Ai}} e^{-k\tau_p N} \right)}$$

where  $N = C_{A0} \cdot C_{B0}$

$$C_{Ai} = \frac{-F + kV_b N + \sqrt{kV_b N - 4kV_b C_{A0} F}}{2kV_b}$$

if  $C_A \neq C_B$  (4)

For T-S model,

$$X_A = 1 + \frac{1}{2C_{A0} k \tau_b} \left( 1 - \sqrt{1 + \frac{4kC_{A0} \tau_b}{kC_{A0} \tau_p + 1}} \right)$$

if  $C_A = C_B$  (5)

$$X_A = 1 + \frac{1}{2k\tau_b C_{A0}} \left[ 1 - k\tau_b N \sqrt{(1 - k\tau_b N)^2 + \frac{4k\tau_b N}{1 + \frac{N - C_{A0}}{C_{A0}} e^{-\tau_p k N}}} \right]$$

if  $C_A \neq C_B$  (6)

The tracer information thus does not give a unique value of conversion for reactions other than the first order. Additional information concerning the relative earliness or lateness of mixing is also necessary.

The following two limits on micromixing were defined by Danckwerts (9):

(a) Complete Segregation: The incoming fluid is broken up into discrete fragments or streaks which are small compared to the tank and uniformly dispersed in it but in which molecules entering together remain together indefinitely.

(b) Maximum Mixedness: The inflowing material is dispersed on the molecular scale in a time much less than the average residence time; the environment of any particular molecules does not tend to contain an excess of molecules which entered at the same time as itself and the mixture is chemically uniform.

From these definitions, it can be seen that micromixing has no effect on plug flow reactors, and material balance for perfect macromixing gives the limit for maximum mixedness for the perfectly stirred tank.

Thus, results from the material balances obtained from T-S and S-T models are limits for maximum mixedness. For the limit of complete segregation, the different streams having different residence times are combined at the reactor outlet to give the average conversion:

$$\langle X_A \rangle = \int_0^{\infty} X_A(\tau) dF(\tau) \quad (7)$$

where  $F(\tau) = 1 - I(\tau)$

For the residence time distribution of the previous section,

$$F(\tau) = 1 - \exp[-(\tau - \tau_p)/\tau_b] U(\tau - \tau_p)$$

where  $U$  is the unit step function,

the conversions of the second-order system for complete segregation are from equation (7) to give:

$$X_A = \frac{kC_{A0}\tau_p}{1 + kC_{A0}\tau_p} + \int_{\tau_p}^{\infty} \frac{kC_{A0}\tau}{1 + kC_{A0}\tau} \cdot \frac{1}{\tau_b} \exp[-(\tau - \tau_p)/\tau_b] \cdot d\tau$$

if  $C_A = C_B$  (8)

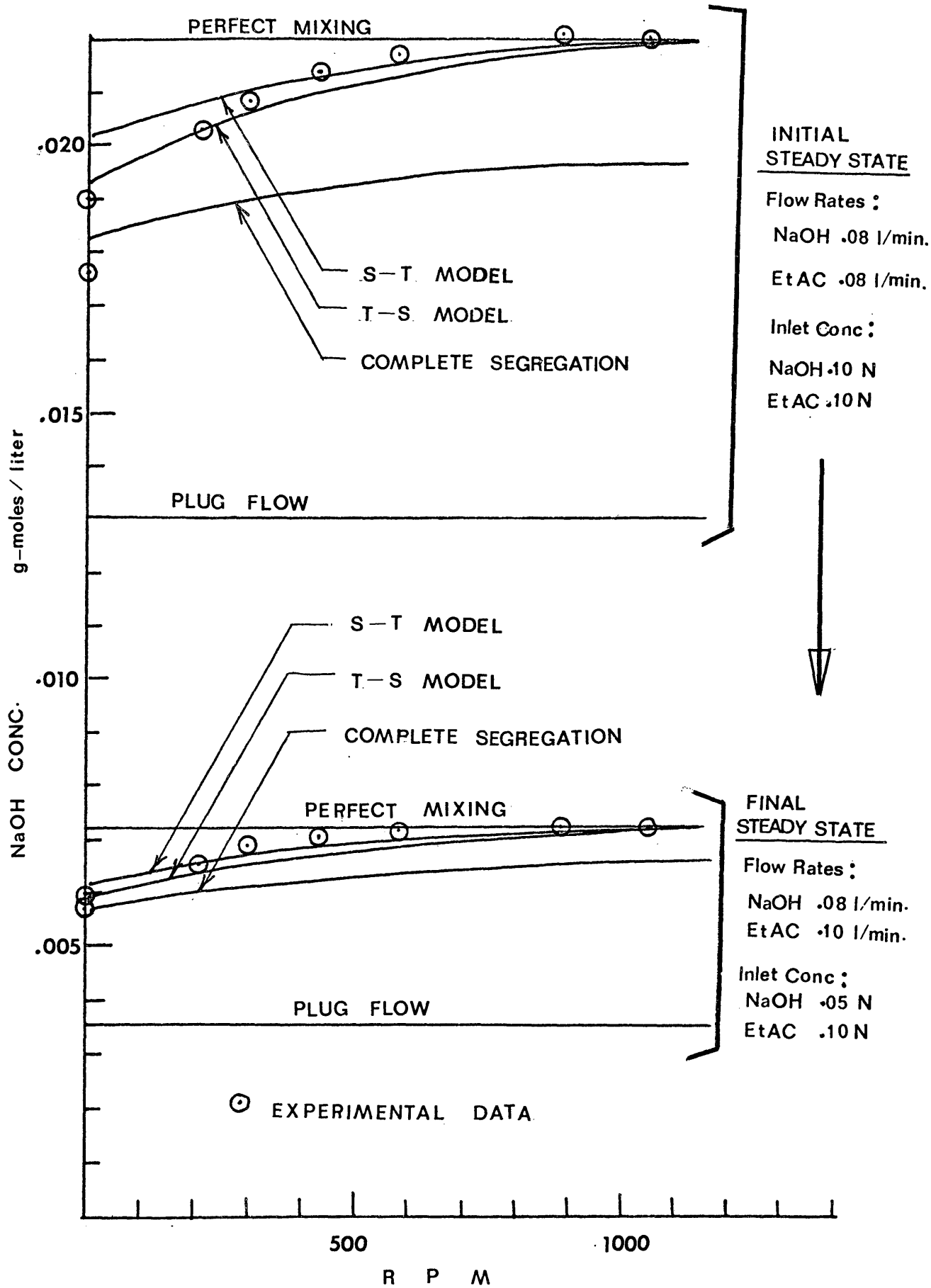
$$X_A = 1 - \frac{N}{C_{A0}(1 + \frac{N-C_{A0}}{C_{A0}} e^{-k\tau_p N})} + \int_{\tau_p}^{\infty} \left\{ 1 - \frac{N}{C_{A0}(1 + \frac{N-C_{A0}}{C_{A0}} e^{-k\tau N})} \right\} \cdot \frac{1}{\tau_b} \exp[-(\tau - \tau_p)/\tau_b] \cdot d\tau$$

if  $C_A \neq C_B$  (9)

Initially, the reactor was operated with the same inlet conditions. The outlet concentration obtained from the experiments as a function of rpm was compared with those of T-S and S-T models and that of complete segregation calculated from equations 5 through 9 in Figure 7.

It appears that macromixing models satisfactorily predict the conversion, although at a very low mixing (lower

Figure 7. Steady state NaOH concentration  
as a function of mixing rate



than approximately 100 rpm) some segregation effect may be present. This result supports the trend indicated by Weinstein and Adler<sup>(21)</sup> that single, isothermal reactions are only slightly affected by micromixing but are substantially affected by macromixing.

It could be concluded from Figure 7 that the limits on conversion will predict performance of the reactor qualitatively. Any inconsistencies shown would be due to errors from age distribution functions which were not exact and to fluctuations in flow rates ( $\pm 3\%$ ).

The rate constant was calculated from the perfectly stirred tank model with the experimental data above 890 rpm, where conversion is not affected with an increase of the mixing rate.

### TRANSIENT CONCENTRATION

In this section, transient concentration of sodium hydroxide to step changes in inlet flow rate of ethyl acetate and inlet concentration of sodium hydroxide is considered at various mixing rates.

The solutions for the ideal cases of perfectly stirred tank and plug flow reactors are obtained by the perturbation method and the method of characteristics, and are combined to give the solution for the mixed models.

#### Perfectly Stirred Tank Reactor

The steady state equations for a second-order system under isothermal condition are:

$$\overline{F}_{A0} \overline{C}_{A0} - \overline{F} \overline{C}_A - kV \overline{C}_A \overline{C}_B = 0 \quad (10a)$$

$$\overline{F}_{B0} \overline{C}_{B0} - \overline{F} \overline{C}_B - kV \overline{C}_A \overline{C}_B = 0 \quad (10b)$$

For the unsteady state, accumulation terms are taken into account to give

$$V \frac{dC_A}{dt} = F_{A0} C_{A0} - F C_A - kV C_A C_B \quad (11a)$$

$$V \frac{dC_B}{dt} = F_{B0} C_{B0} - F C_B - kV C_A C_B \quad (11b)$$

For step changes in inlet concentration of sodium hydroxide (A) by  $\alpha$  and inlet flow rate of ethyl acetate (B) by  $\beta$ , each variable can be written as the sum of the steady state value and the perturbed value as follows:

$$C_{Ao} = \bar{C}_{Ao} + \alpha$$

$$C_A = \bar{C}_A + C'_A$$

$$C_B = \bar{C}_B + C'_B$$

$$F_{Bo} = \bar{F}_{Bo} + \beta$$

$$F = \bar{F} + \beta$$

Substituting these into the unsteady state equations and subtracting the steady state equations, we get

$$\frac{dC'_A}{dt} = a_1 + b_1 C'_A + c_1 C'_B + d C'_A C'_B \quad (12a)$$

$$\frac{dC'_B}{dt} = a_2 + b_2 C'_A - c_2 C'_B + d C'_A C'_B \quad (12b)$$

where

$$a_1 = (\alpha F_{Ao} - \beta \bar{C}_A) / V \quad a_2 = (\beta C_{Bo} - \beta C_{Bo}) / V$$

$$b_1 = -(\bar{F} + \beta + kV\bar{C}_B) / V \quad b_2 = -k\bar{C}_B$$

$$c_1 = -k\bar{C}_A \quad c_2 = -(\bar{F} + \beta + kV\bar{C}_A) / V$$

$$d = -k$$

(For convenience, primes will be deleted henceforth.)

The system of equations (12a, b) can be easily solved by the Laplace transform method or characteristic root method, if the flow rates and concentrations are such that  $C_A$  can be

written as a function of  $C_B$  (e.g.  $C_A = C_B$ , and  $F_{A0} = F_{B0}$ ) or if the perturbation is small and the nonlinear terms are negligible. However, when the nonlinear terms are not negligible, as in the present case, they should be taken into account.

Numerical methods, such as the predictor-corrector method or the Runge-Kutta method, are at hand, but they require considerable time for derivative evaluations and iterations. Following is the solution of the system of equations 12a, 12b by the perturbation method.

First, nonlinear terms are perturbed by the perturbation parameter  $\mu$ , where  $\mu < 1$ :

$$\frac{dC_A}{dt} = a_1 + b_1 C_A + c_1 C_B + \mu d C_A C_B \quad (13a)$$

$$\frac{dC_B}{dt} = a_2 + b_2 C_A + c_2 C_B + \mu d C_A C_B \quad (13b)$$

Then the transient concentrations are sought in terms of a power series in  $\mu$  as follows:

$$C_A = C_{A0}(t) + \sum_{i=1}^n \mu^i C_{Ai}(t) \quad (14a)$$

$$C_B = C_{B0}(t) + \sum_{i=1}^n \mu^i C_{Bi}(t) \quad (14b)$$

For the first order approximation, substitute

$$C_A(t) = C_{A0}(t) + \mu C_{A1}(t)$$

$$C_B(t) = C_{B0}(t) + \mu C_{B1}(t)$$

into equations 13a and 13b to give

$$\begin{aligned} \frac{dC_{A0}}{dt} + \mu \frac{dC_{A1}}{dt} &= a_1 - b_1 C_{A0} + c_1 C_{B0} \\ &+ \mu (b_1 C_{A1} - c_1 C_{B1} + d C_{A0} C_{B0}) \\ &+ \text{higher order terms in } \mu \end{aligned} \quad (15a)$$

$$\begin{aligned} \frac{dC_{B0}}{dt} + \mu \frac{dC_{B1}}{dt} &= a_2 - b_2 C_{A0} + c_2 C_{B0} \\ &+ \mu (b_2 C_{A1} - c_2 C_{B1} + d C_{A0} C_{B0}) \\ &+ \text{higher order terms in } \mu. \end{aligned} \quad (15b)$$

Matching the coefficients of  $\mu^0$  and  $\mu^1$ ,

we get

$$\frac{dC_{A0}}{dt} = a_1 + b_1 C_{A0} + c_1 C_{B0} \quad (16a)$$

$$\frac{dC_{B0}}{dt} = a_2 + b_2 C_{A0} + c_2 C_{B0} \quad (16b)$$

$$\frac{dC_{A1}}{dt} = b_1 C_{A1} + c_1 C_{B1} + d C_{A0} C_{B0} \quad (17a)$$

$$\frac{dC_{B1}}{dt} = b_2 C_{A1} + c_2 C_{B1} + d C_{A0} C_{B0} \quad (17b)$$

These systems of equations are linear and are solved as follows.

First we solve the system of equations 16a and 16b. Taking the Laplace transformation, with the initial conditions

$$C_{A0}(0) = C_{B0}(0) = 0,$$

and rearranging, we get,

$$(s-b_1)\bar{C}_{Ao} + c_1\bar{C}_{Bo} = a_1/s \quad (18a)$$

$$-b_2\bar{C}_{Ao} + (s-c_2)\bar{C}_{Bo} = a_2/s \quad (18b)$$

Solving for  $\bar{C}_{Ao}$  and  $\bar{C}_{Bo}$

$$\bar{C}_{Ao} = \frac{a_1(s-\alpha_3)}{s(s-\alpha_1)(s-\alpha_2)} \quad (19a)$$

$$\bar{C}_{Bo} = \frac{a_2(s-\alpha_4)}{s(s-\alpha_1)(s-\alpha_2)} \quad (19b)$$

where

$$\alpha_1 = \frac{1}{2} [b_1 + c_2 + \sqrt{(b_1+c_2)^2 - 4(b_1c_2 - b_2c_1)}]$$

$$\alpha_2 = \frac{1}{2} [b_1 + c_2 - \sqrt{(b_1+c_2)^2 - 4(b_1c_2 - b_2c_1)}]$$

$$\alpha_3 = c_2 + a_2c_1/a_1$$

$$\alpha_4 = b_1 + a_1b_2/a_2$$

Inverting equations 19a and 19b, we get

$$C_{Ao} = \beta_0 [1 + (\beta_1 e^{\alpha_1 t} + \beta_2 e^{\alpha_2 t})] \quad (20a)$$

$$C_{Bo} = \beta_3 [1 + (\beta_4 e^{\alpha_1 t} + \beta_5 e^{\alpha_2 t})] \quad (20b)$$

where

$$\beta_0 = -\frac{a_1\alpha_3}{\alpha_1\alpha_2}$$

$$\beta_1 = \frac{a_1(\alpha_3 - \alpha_1)}{\alpha_1(\alpha_1 - \alpha_2)\beta_0}$$

$$\beta_2 = \frac{a_1(\alpha_3 - \alpha_2)}{\alpha_2(\alpha_2 - \alpha_1)\beta_0}$$

$$\beta_1 + \beta_2 = 1$$

and

$$\beta_3 = \dots \frac{a_2 \alpha_4}{\alpha_1 \alpha_2}$$

$$\beta_4 = \frac{a_2 (\alpha_4 - \alpha_1)}{1 (\alpha_1 - \alpha_2) \beta_3}$$

$$\beta_5 = \frac{a_2 (\alpha_4 - \alpha_2)}{\alpha_2 (\alpha_2 - \alpha_1) \beta_3}$$

$$\beta_5 + \beta_4 = 1$$

$C_{A0}(t)$  and  $C_{B0}(t)$  (equations 20a and 20b) are the solutions for the small perturbations.

We proceed to solve the first-order correction terms,  $C_{A1}$  and  $C_{B1}$ , by substituting  $C_{A0}$  and  $C_{B0}$  (equations 20a and 20b) to equations 17a and 17b and solving the resulting system of equations.

$$\begin{aligned} \frac{dC_{A1}}{dt} = & b_1 C_{A1} + c_1 C_{B1} \\ & + d \beta_0 \beta_3 \left[ 1 \quad (\beta_1 e^{\alpha_1 t} + \beta_2 e^{\alpha_2 t}) \right] \left[ 1 \quad (\beta_4 e^{\alpha_1 t} + \beta_5 e^{\alpha_2 t}) \right] \end{aligned} \quad (21a)$$

$$\begin{aligned} \frac{dC_{B1}}{dt} = & b_2 C_{A1} + c_2 C_{B1} \\ & + d \beta_0 \beta_3 \left[ 1 \quad (\beta_1 e^{\alpha_1 t} + \beta_2 e^{\alpha_2 t}) \right] \left[ 1 \quad (\beta_4 e^{\alpha_1 t} + \beta_5 e^{\alpha_2 t}) \right] \end{aligned} \quad (21b)$$

Taking the Laplace transformation of equations 21a and 21b with the initial conditions

$$C_{A1}(0) = C_{B1}(0) = 0$$

and proceeding as before, we get

$$\begin{aligned}
c_{A1} = \alpha\beta_0\beta_3 & \left\{ \frac{c_1 - c_2}{\alpha_1\alpha_2} \right. \\
& \cdot e^{\alpha_1 t} \left[ \frac{c_1 - c_2 + \alpha_1}{\alpha_1(-\alpha_1 + \alpha_2)} + (\beta_1 + \beta_4) \left( \frac{c_1 - c_2 + \alpha_1}{-\alpha_2 + \alpha_1} t + \frac{c_2 - c_1 - \alpha_2}{(\alpha_2 + \alpha_1)^2} \right) + \frac{(\beta_2 + \beta_5)(c_1 - c_2 + \alpha_1)}{(-\alpha_1 + \alpha_2)^2} \right] \\
& \cdot \frac{(\beta_2\beta_5)(c_1 - c_2 + \alpha_1)}{(\alpha_1 - \alpha_2)(\alpha_1 + 2\alpha_2)} - \frac{\beta_1\beta_4(c_1 - c_2 + \alpha_1)}{(-\alpha_2 + \alpha_1)(-\alpha_1)} \cdot \frac{(\beta_1\beta_5 + \beta_2\beta_4)(c_1 - c_2 + \alpha_1)}{(\alpha_1 + \alpha_2)(-\alpha_2)} \left. \right\} \\
& \cdot \alpha_2 t \left[ \frac{c_1 - c_2 + \alpha_2}{\alpha_2(\alpha_1 - \alpha_2)} + \frac{(\beta_1 + \beta_4)(c_1 - c_2 + \alpha_2)}{(\alpha_1 - \alpha_2)^2} + (\beta_2 + \beta_5) \left( \frac{c_2 - c_1 - \alpha_2}{(-\alpha_1 + \alpha_2)} t + \frac{c_2 - c_1 - \alpha_1}{(-\alpha_1 + \alpha_2)^2} \right) \right] \\
& + \beta_2\beta_5 \frac{(-c_2 + c_1 + \alpha_2)}{(-\alpha_1 + \alpha_2)\alpha_2} - \frac{\beta_1\beta_4(-c_2 + c_1 + \alpha_2)}{(-\alpha_1 + \alpha_2)(-2\alpha_1 + \alpha_2)} + \frac{(\beta_1\beta_5 + \beta_2\beta_4)(-c_2 + c_1 + \alpha_2)}{(-\alpha_1 + \alpha_2)\alpha_1} \left. \right\} \\
& \cdot e^{2\alpha_1 t} \left[ \frac{-\beta_1\beta_4(-c_2 + c_1 + 2\alpha_1)}{\alpha_1(-\alpha_2 + 2\alpha_1)} \right] \\
& \cdot e^{2\alpha_2 t} \left[ \frac{-\beta_2\beta_5(-c_2 + c_1 + 2\alpha_2)}{2(-\alpha_1 + 2\alpha_2)} \right] \\
& \cdot e^{(\alpha_1 + \alpha_2)t} \left[ \frac{-(\beta_1\beta_5 + \beta_2\beta_4)(-c_2 + c_1 + \alpha_1 + \alpha_2)}{\alpha_1\alpha_2} \right] \left. \right\} \quad (22)
\end{aligned}$$

The assumption that

$$\left[ 1 - (\beta_1 e^{\alpha_1 t} + \beta_2 e^{\alpha_2 t}) \right] \left[ 1 - (\beta_4 e^{\alpha_1 t} + \beta_5 e^{\alpha_2 t}) \right] \doteq 1 - e^{\alpha t} \quad (23)$$

where  $\alpha = \frac{1}{2} (\alpha_1 + \alpha_2)$

gives simplified solutions for the first-order correction terms,

$$c_{A1} = d\beta_0\beta_3 (\gamma_0 - \gamma_1 e^{\alpha_1 t} - \gamma_2 e^{\alpha_2 t} - \gamma_3 e^{\alpha t}) \quad (23a)$$

where

$$\gamma_0 = \frac{c_1 - c_2}{\alpha_1 \alpha_2}$$

$$\gamma_1 = \frac{(c_1 - c_2 + 1) \alpha}{\alpha_1 (\alpha_2 - \alpha_1) (\alpha - \alpha_1)}$$

$$\gamma_2 = \frac{(c_1 - c_2 + \alpha_2) \alpha}{\alpha_2 (\alpha_1 - \alpha_2) (\alpha - \alpha_2)}$$

$$\gamma_3 = \frac{c_1 - c_2 + \alpha}{(\alpha - \alpha_1) (\alpha - \alpha_2)}$$

$$c_{B1} = d\beta_0\beta_3 (\gamma_4 - \gamma_5 e^{\alpha_1 t} - \gamma_6 e^{\alpha_2 t} - \gamma_7 e^{\alpha t}) \quad (23b)$$

where

$$\gamma_4 = \frac{b_2 - b_1}{\alpha_1 \alpha_2}$$

$$\gamma_5 = \frac{\alpha (b_2 - b_1 + \alpha_1)}{\alpha_1 (\alpha_2 - \alpha_1) (\alpha - \alpha_1)}$$

$$\gamma_6 = \frac{\alpha (b_2 - b_1 + \alpha_2)}{\alpha_2 (\alpha_1 - \alpha_2) (\alpha - \alpha_2)}$$

$$\gamma_7 = \frac{b_2 - b_1 + \alpha}{(\alpha - \alpha_1) (\alpha - \alpha_2)}$$

For the present experimental setup with large concentration and flow rate upsets, transient sodium hydroxide concentration calculated through the first-order correction terms using equation 22 showed little difference from that using equation 23a, as the following table shows, and the assumption of equation 23 appears to be good.

TABLE Transient NaOH Concentration

<u>Time (min.)</u>	<u>using Eq.22</u>	<u>using Eq. 23a</u>
0	0.02200	0.02200
1	0.01947	0.01916
2	0.01722	0.01697
3	0.01552	0.01572
7	0.01055	0.01108
10	0.00888	0.00856
12	0.00826	0.00799
$\infty$	0.00716	0.00716

Continuing with the approximations

$$C_{Ai}C_{Bj} \doteq C_{A,n-1}C_{B,n-1} \quad i, j \leq n-1 \quad (24a)$$

$$C_{An} \doteq C_{A,n-1}(\infty)(1 - e^{\alpha t}) \quad (24b)$$

$$C_{Bn} \doteq C_{B,n-1}(\infty)(1 - e^{\alpha t}) \quad (24c)$$

we get the following general expressions for the correction terms:

$$C_{An} = n d C_{A,n-1}(\infty) C_{B,n-1}(\infty) (\gamma_0 - \gamma_1 e^{\alpha_1 t} - \gamma_2 e^{\alpha_2 t} - \gamma_3 e^{\alpha t}) \quad (25a)$$

$$C_{Bn} = n d C_{A,n-1}(\infty) C_{B,n-1}(\infty) (\gamma_4 - \gamma_5 e^{\alpha_1 t} - \gamma_6 e^{\alpha_2 t} - \gamma_7 e^{\alpha t}) \quad (25b)$$

Now convergence of the correction terms can be easily checked by equations 24a and 24b. Then we relax  $\mu$  to 1 to get the transient concentrations

$$C_A = \bar{C}_A + C_{A0} + C_{A1} + \dots \quad (26a)$$

$$C_B = \bar{C}_B + C_{B0} + C_{B1} + \dots \quad (26b)$$

For the present experimental setup, the transient concentration converged after the first correction terms to 0.5%.

### Plug Flow Reactor

The unsteady state equation for the second-order PF reactor is

$$\frac{\partial C_A}{\partial t} + v \frac{\partial C_A}{\partial z} = -k C_A C_B \quad (27)$$

For the inlet flow upset, we write linear flow velocity as the sum of steady state value and perturbation.

$$v = \bar{v} + v'$$

With the change of variables,

$$\theta = vt/L; \quad y = z/L; \quad r = v'/\bar{v}; \quad X = C_A/C_{A0}$$

equation 27 becomes

$$\frac{\partial X}{\partial \theta} + (1 - r) \frac{\partial X}{\partial y} = -\frac{kL}{\bar{v}} X C_B \quad (28)$$

Substituting the stoichiometric relation

$$C_B = C_A - (C_{A0} - C_{B0})$$

into equation 28, we get

$$\begin{aligned} \frac{\partial X}{\partial \theta} + \left(1 + \frac{v'}{\bar{v}}\right) \frac{\partial X}{\partial y} &= - \frac{kL}{\bar{v}} X C_A - (C_{A0} - C_{B0}) \\ &= - \frac{kL C_{A0}}{\bar{v}} X(X - M) \\ &= - P X(X - M) \end{aligned} \quad (29)$$

where  $M = (C_{A0} - C_{B0})/C_{A0}$  and  $P = kL C_{A0} / \bar{v}$

The change of variable suggested by Koppel<sup>(22)</sup>

$$\varphi = \begin{cases} - \frac{1}{P} \int_c^X \frac{d\xi}{\xi(\xi - M)} = \frac{1}{PM} \ln \frac{X(C-M)}{C(X-M)} & \text{for } M \neq 0 \\ - \frac{1}{P} \int_c^X \frac{d\xi}{\xi^2} = \frac{1}{PX} - C & \text{for } M=0 \end{cases} \quad (30)$$

where C is an arbitrary constant,

reduces equation (30) to

$$\frac{\partial \varphi}{\partial \theta} + [1 + r(\theta)] \frac{\partial \varphi}{\partial y} = 1 \quad \text{where } r(\theta) = v'/\bar{v} \quad (31)$$

Equation 31 is written as two ordinary differential equations by the method of characteristics,

$$d\theta = \frac{dy}{1 + r(\theta)} = d\varphi \quad (32)$$

$$d\theta = d\varphi \quad (33)$$

The solutions of these are

$$\theta + \int_0^\theta r(\tau) d\tau - y = K_1 \quad (34)$$

$$\theta - \varphi = K_2 \quad (35)$$

Where  $K_1$  and  $K_2$  are arbitrary constants.

The solution of equation 31 was obtained by the method of characteristics for the arbitrary initial conditions and forcing functions by Koppel.<sup>(22)</sup> Following is the solution procedure for step changes in inlet concentration of A and inlet flow rate of B by the method of characteristics.

The general solution of equation 31 is  $K_2 = f(K_1)$ , where  $f$  is an arbitrary function.

Thus,

$$\theta - \varphi = f\left(\theta + \int_0^\theta r(\tau) d\tau - y\right) \quad (36)$$

For the step change in inlet concentration of A by  $\alpha$ ,

$$\varphi(0, \theta) = \varphi(0, 0) + \alpha U(\theta)$$

where  $U(\theta)$  is the unit step function and  $\varphi(0, 0)$  is the initial steady state value.

This condition reduces equation 36 to

$$\theta - \varphi(0, 0) - \alpha U(\theta) = f\left(\theta + \int_0^\theta r(\tau) d\tau\right) \quad (37)$$

Equation 37 is the functional relation being sought.

To solve this, set

$$z = \theta + \int_0^\theta r(\tau) d\tau \quad (38)$$

$$\theta = z - \int_0^{g(z)} r(\tau) d\tau \quad (39)$$

where  $g(z)$  is a function satisfying

$$g(z) = z - \int_0^{g(z)} r(\tau) d\tau \quad (40)$$

The functional relation  $f$  can be obtained by substituting equations 38 and 39 into equation 37.

$$f(z) = z \cdot \int_0^{g(z)} r(\tau) d\tau - \varphi(0,0) - \alpha U(\theta) \quad (41)$$

Now that the function  $f$  is known, it can be used in equation 36 to give

$$\varphi = y \cdot \int_T^\theta r(\tau) d\tau + \varphi(0,0) + \alpha U(z - \int_0^{g(z)} r(\tau) d\tau) \quad (42)$$

where  $T = g(\theta + \int_0^\theta r(\tau) d\tau - y)$

For the step change in inflow rate by  $\beta$

$$r(\theta) = \beta U(\theta) \quad (43)$$

Then,

$$g(y) = y \cdot \beta g(y) \quad (44)$$

$$g(y) = y/(1 + \beta) \quad (44)$$

$$g(\theta + \beta\theta - y) = \theta \cdot y/(1 + \beta) \quad (45)$$

The solution for the transient concentration of A then becomes

$$\varphi(y,0) = \begin{cases} y \cdot (\beta\theta + \varphi_0(0,0)) & \text{for } \theta < \frac{y}{1+\beta} \\ \frac{y}{1+\beta} + \varphi_1(0,0) & \text{for } \theta > \frac{y}{1+\beta} \end{cases} \quad (46)$$

where  $\varphi_0(0,0)$  is the concentration of A before the upset at the inlet in terms of  $\varphi$  and  $\varphi_1(0,0)$  is that after the upset.

For the transient concentration at the outlet,  $y = 1$ , and equation 35 becomes

$$\varphi(1, \theta) = \begin{cases} 1 - \beta\theta + \varphi_0(0,0) & \text{for } \theta < \frac{1}{1+\beta} \\ \frac{1}{1+\beta} + \varphi_1(0,0) & \text{for } \theta > \frac{1}{1+\beta} \end{cases} \quad (47)$$

The transient sodium hydroxide concentrations for the perfectly stirred tank by equation 25a and the plug flow reactor by equation 47 are shown in Figures 8a through 8c. Note that for the plug flow reactor, inlet concentration change is not felt until it reaches the outlet, whereas inlet flow rate increase changes the output concentration due to change in residence time. Combining these two with appropriate changes in initial conditions gives the expression for the mixed models. For T-S models, the inlet concentration and flow rate upsets were hardly felt at the exit of plug flow portion until one residence time because the plug flow portion was at most approximately 8% at no mixing. Thereafter the outlet concentration showed step changes for the perfectly stirred tank portion. The resulting solutions were compared with the experimental data in Figures 8a through 8d. Note that the curve for the T-S models are based on the residence time distribution functions for the lower rpm given in each of these figures. At no mixing (Figure 8a) the experimental curves were somewhat lower than those for T-S models, indicating segregation may be present. As the mixing increases the T-S models seem to get closer to the experimental data, indicating

Figure 8a. Transient NaOH concentration  
at no mixing

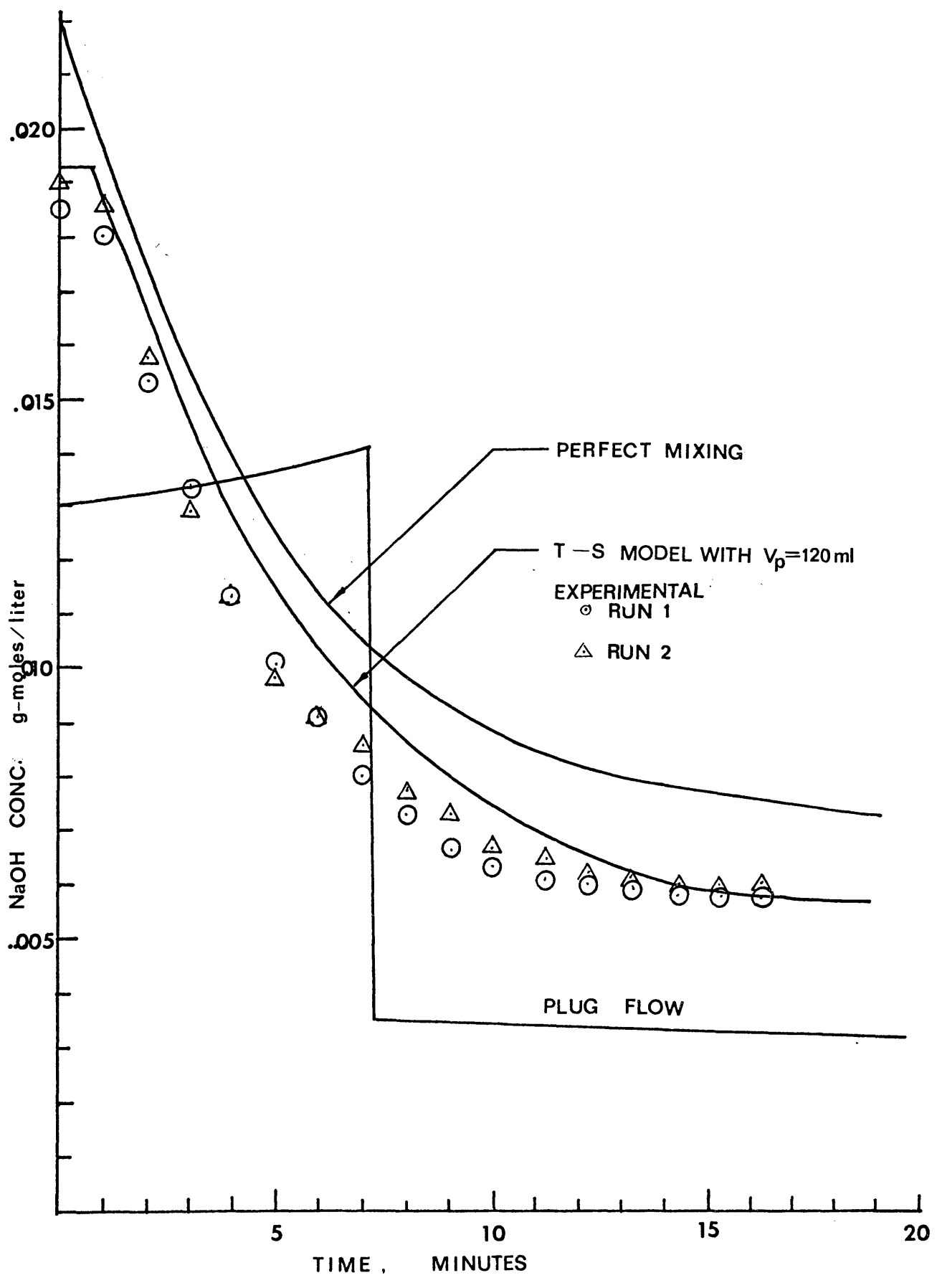


Figure 8b. Transient NaOH concentration  
at 205 and 300 rpm.

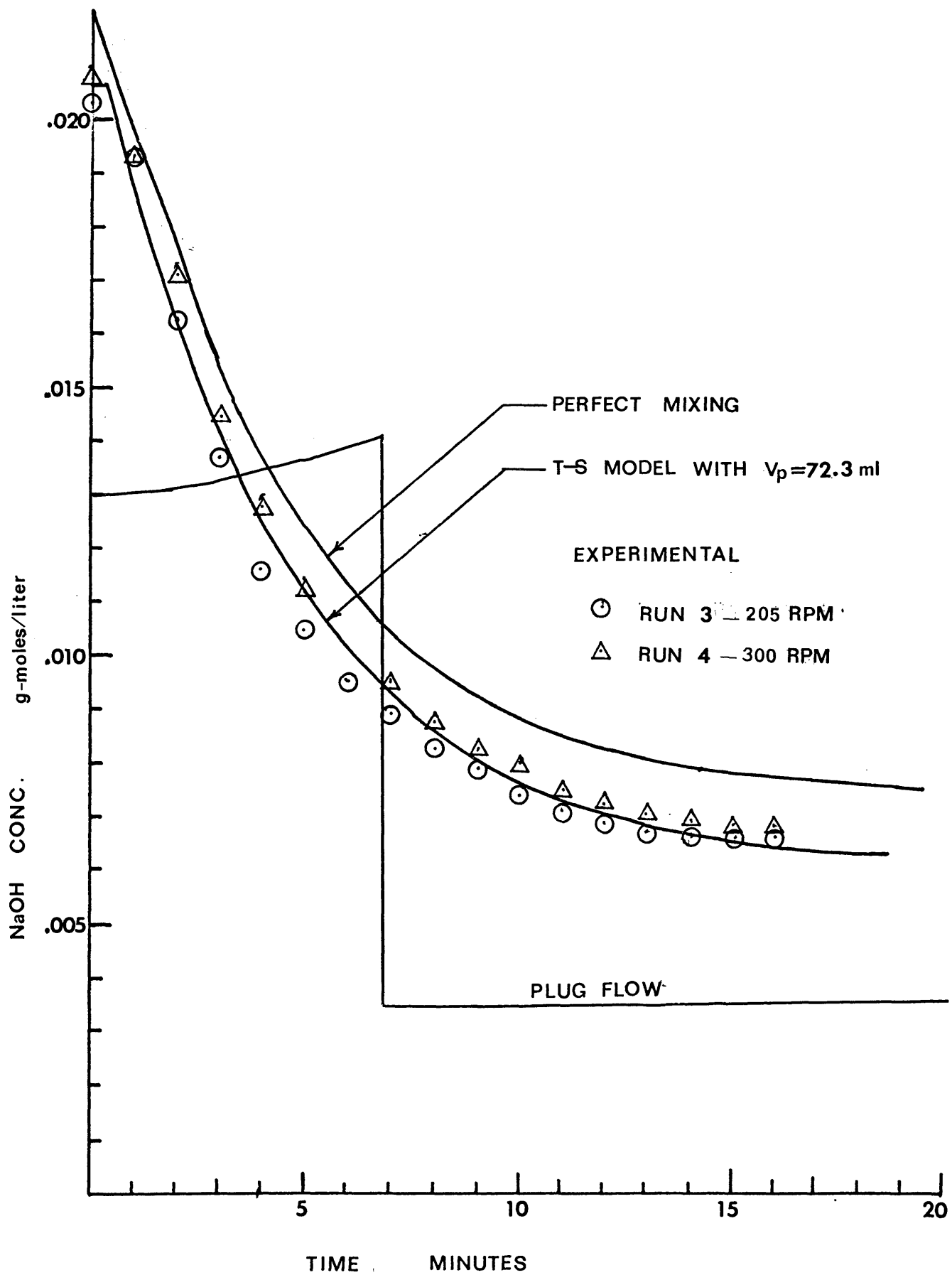


Figure 8c. Transien NaOH concentration  
at 433 and 582 rpm.

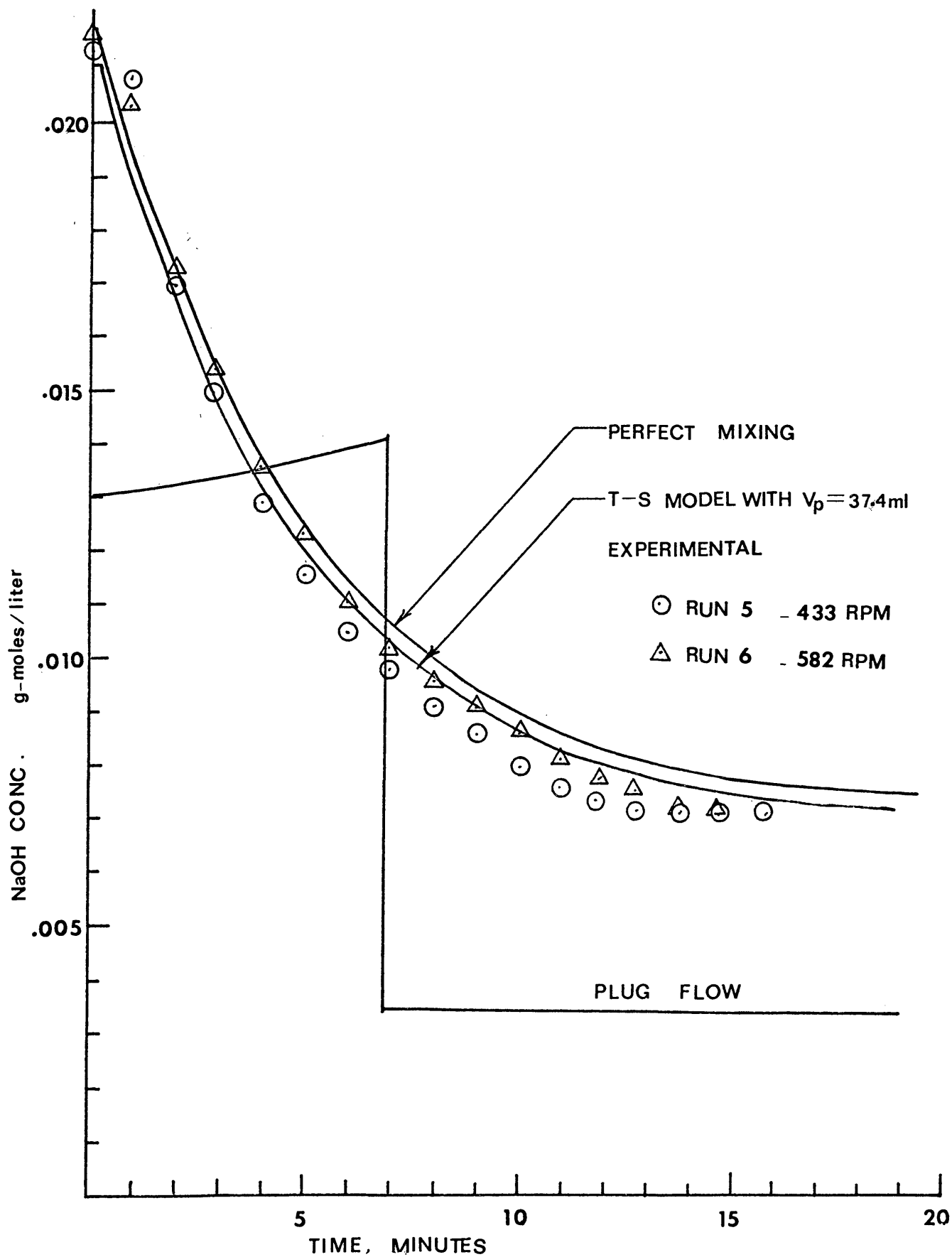
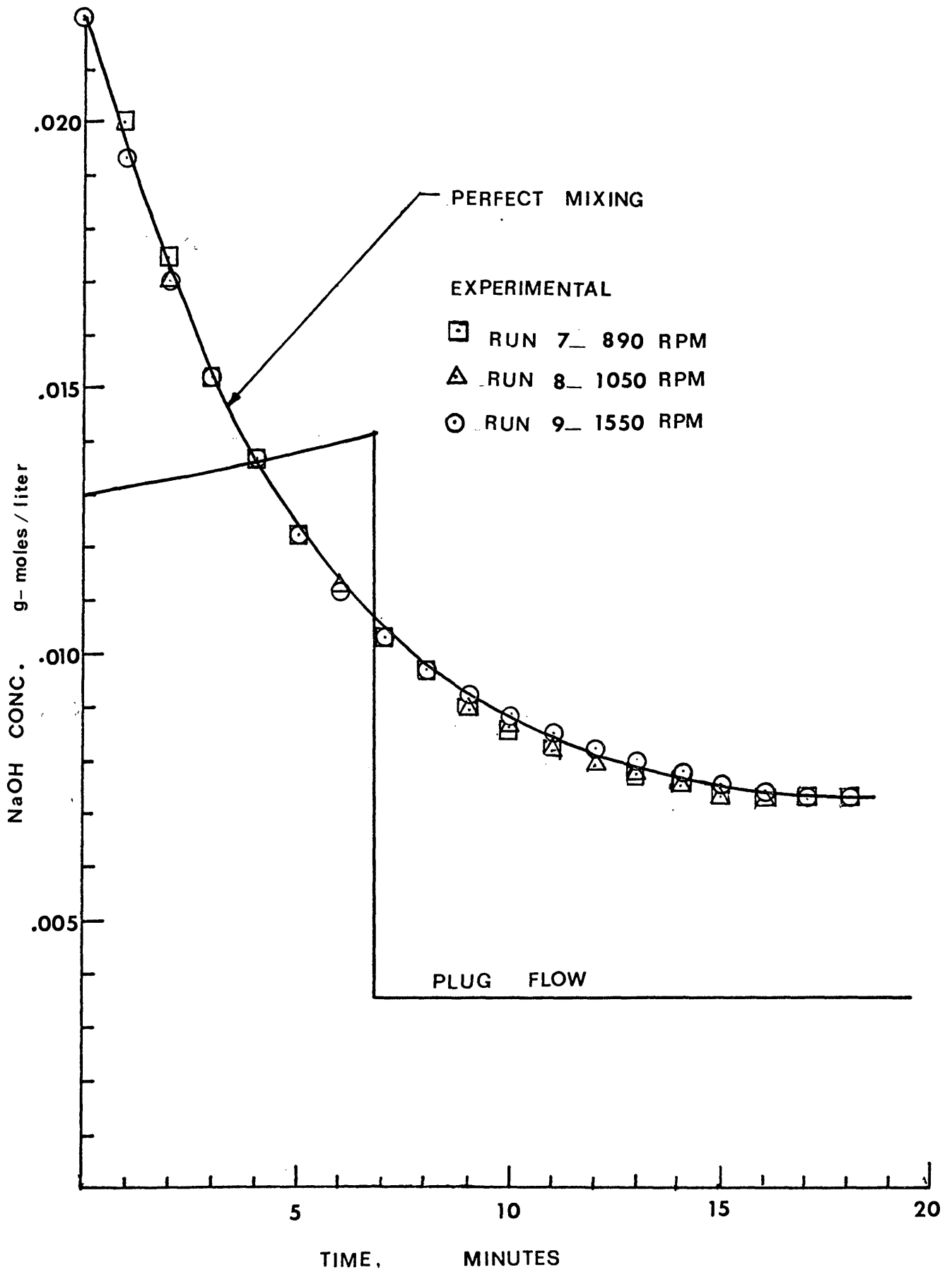


Figure 8d. Transient NaOH concentration  
at 890, 1050, 1550 rpm.

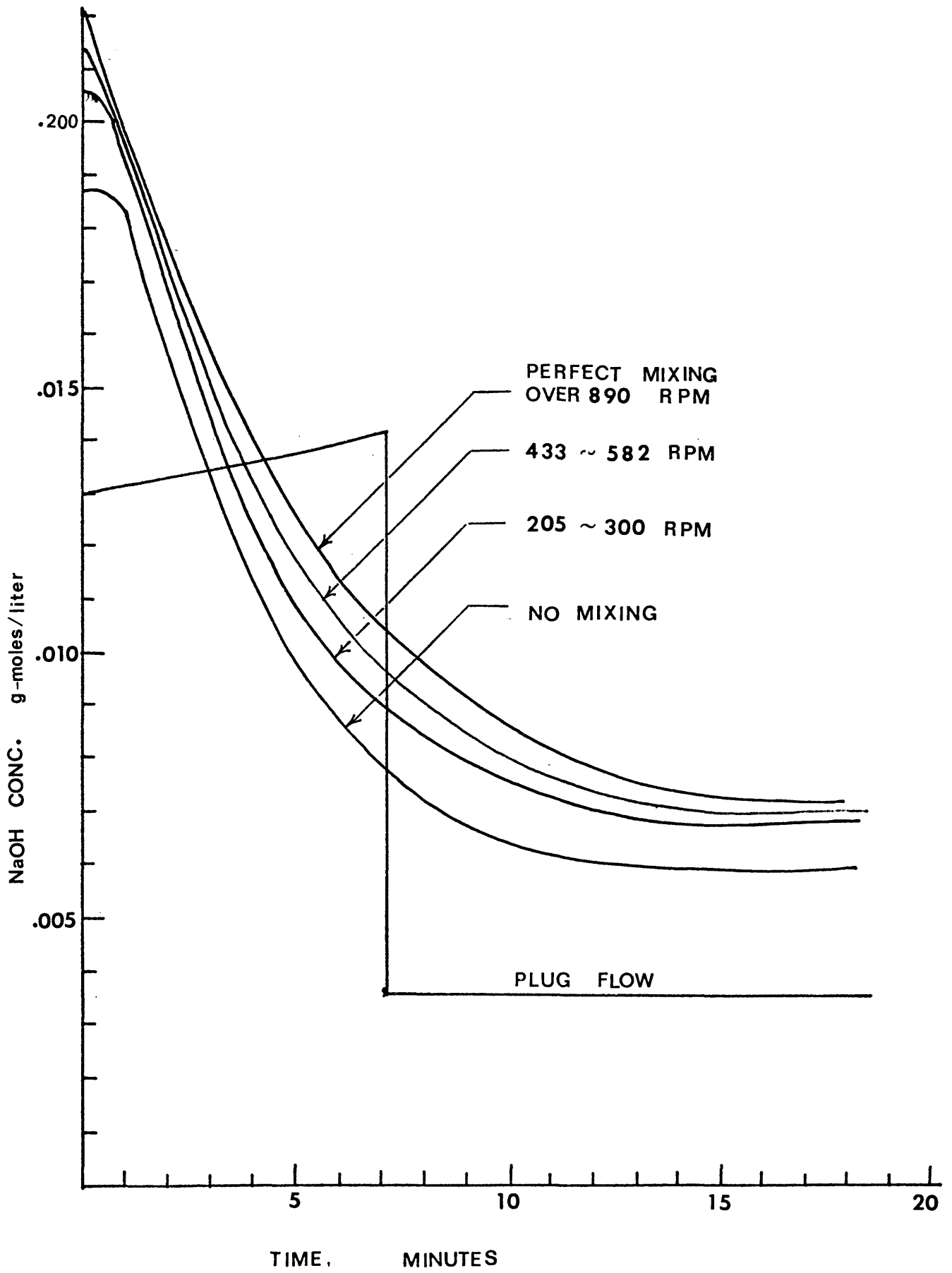


macromixing information is sufficient for the description of the reactor performance.

The large portion of the curves show exponential change, which confirms the tracer results that a large fraction of the reactor is perfectly mixed. The existence of a delay in response close to time zero shows the effect of plug flow. From these facts, transient runs for a reacting system could be used directly to qualitatively describe the performance of a reactor and replace the tracer run for more realistic determination of the residence time distribution.

Figure 8e show the trend that conversion decreases with the increase of the mixing rate due to the decrease of plug flow portion as evidenced by the delay in response near time zero.

Figure 8e. Comparison of the experimental transient NaOH concentration at various rpm range.



### CONCLUSIONS

For the reaction system adopted in this study (second order, isothermal), a plug flow reactor and a perfectly stirred tank reactor in series were found as appropriate models for macromixing by tracer experiments. It was found that the plug flow portion exponentially decreased with the increase of the mixing rate.

By comparing the conversions at various mixing rates with those of complete segregation, it was found that, at a very low mixing rate (close to no mixing), there existed a small effect of segregation. In general, the micromixing effect was much smaller than that of macromixing.

The perturbation method was found to be time saving for calculations for the transient concentration of the perfectly mixed reactor, and the method of characteristics of Koppel was extended for the solution of the transient concentration of the plug flow reactor.

Comparison of the experimental transient concentration with those of the mixed model predicted by the tracer runs showed good agreement. This suggests that the transient runs for a real reacting system could be an effective method

to observe the nonideal behavior of imperfectly mixed reactors.

It was shown for the second-order reaction that conversion decreased with the increase of the mixing rate. This result does not necessarily suggest that we could leave the reactor unmixed for the maximum conversion, because there existed higher noise at the lower mixing rate, which might cause difficulties with reactor control.

Mixing pattern is a strong function of the reactor geometry. The theory developed herein can be applied to other reactor geometries by obtaining the tracer response of the system in question. The model then allows prediction of the reaction conversion for this new system.

LIST OF NOMENCLATURE

- C: Concentration (g-mole/liter)
- F: Volumetric flow rate (liter/min)
- $\underline{F}$ : Residence time distribution function,  $1 - C_A(t)/C_{A0}$
- $\underline{I}$ : Residence time distribution function,  $1 - \underline{F}$
- k: Second-order rate constant (liter/mole min)
- L: Reactor length (ft)
- M:  $(C_{A0} - C_{B0})/C_{A0}$
- N:  $C_{A0} - C_{B0}$
- P:  $kLC_{A0}/\bar{v}$
- U: Unit step function
- V: Reactor volume (liter)
- v: Linear flow rate (ft/min)
- X: Conversion,  $1 - C_A/C_{A0}$
- $\alpha$ : Size of the step change in inlet NaOH concentration
- $\beta$ : Size of the step change in inlet EtAc flow rate
- $\alpha_i$ : Constants
- $\beta_i$ : Constants
- $\gamma_i$ : Constants

- y: Dimensionless distance  
r(t): Dimensionless perturbation of velocity  
 $\mu$ : Perturbation parameter for the perturbation method  
 $\varphi$ : Transformed dimensionless concentraion defined by equation 30.  
 $\tau$ : V/F  
 $\xi$ : Dummy variable of integration

### Subscripts

- A: Sodium hydroxide  
B: Ethyl acetate  
b: Perfect backmixing  
p: Plug flow  
(i): i-th correction term in the perturbation method

### Superscripts

- $\bar{\phantom{x}}$ : Steady state  
 $\bar{\phantom{x}}^{\infty}$ : Laplace transformation  
 $\prime$ : Perturbation

APPENDIXEXPERIMENTAL DATATracer Runs

A: NaOH            Temperature: 80°F

Initially  $C_A = 0$ , Reactor volume=1225 ml

At time zero, upset  $C_A$  to 0.1 g-moles/liter

Flow Rates 0.16 liter/minute

Run 1. No mixing

<u>time (min)</u>	<u>0</u>	<u><math>C_A</math></u>	<u>I</u>
0.65	0.0675	0.	1.
1.0	0.1306	0.0026	0.974
1.5	0.1959	0.0100	0.900
3.0	0.3918	0.0263	0.737
5.0	0.6530	0.0247	0.573
7.65	1.	0.0617	0.383
9.0	1.1754	0.0692	0.308
11.0	1.4366	0.0776	0.224
13.0	1.6978	0.0832	0.168
15.3	2.	0.0871	0.139

Run 2. No mixing

<u>time (min)</u>	<u>0</u>	<u>C<sub>A</sub> (mol/l)</u>	<u>I</u>
0.65	0.0675	0.0000	1.000
1.0	0.1306	0.0000	1.000
1.5	0.1959	0.0007	0.993
2.0	0.2612	0.0186	0.814
4.0	0.5224	0.0331	0.669
6.0	0.7856	0.0537	0.463
8.0	1.0448	0.0708	0.292
10.0	1.3060	0.0794	0.206

Run 3. 194 rpm

0.43	0.056	0.0000	1.000
1.0	0.1356	0.0069	0.931
2.0	0.2612	0.0126	0.874
3.0	0.3918	0.0224	0.776
4.0	0.5224	0.0324	0.676
5.0	0.6530	0.0437	0.563
7.65	1.0000	0.0676	0.324
10.0	1.3060	0.0759	0.241

Run 4. 279 rpm

0.25	0.0327	0.0000	1.000
1.53	0.2	0.0282	0.718
3.06	0.4	0.0437	0.563
6.12	0.8	0.0550	0.450
7.65	1.0	0.0646	0.354
9.18	1.2	0.0724	0.276
10.7	1.4	0.0776	0.224
12.5	1.6	0.0893	0.107

Run 5. 413 rpm

<u>Time (min)</u>	<u>0</u>	<u>C<sub>A</sub>(mole/l)</u>	<u>I</u>
0.313	0.0410	0.0000	1.000
1.5	0.1959	0.0149	0.851
3.0	0.3918	0.0277	0.723
5.0	0.6530	0.0464	0.536
7.65	1.0	0.0650	0.350
9.0	1.1754	0.0702	0.298
11.0	1.4366	0.0775	0.225
13.0	1.6979	0.0827	0.173
15.3	2.0	0.0873	0.127

Run 6. 615 rpm (Time lag=0.10 min.)Run 7. 1060 rpm (Time lag=0.05 min.)Run 8. 1290 rpm (Time lag=0.)

1.5	0.1959	0.0173	0.827
3.0	0.3918	0.0323	0.677
5.0	0.6530	0.0480	0.520
7.65	1.0	0.0632	0.368
9.0	1.1754	0.0692	0.308
11.0	1.4366	0.0763	0.237
13.0	1.6979	0.0817	0.183
15.3	2.0	0.0865	0.135

Transient Reaction Runs

Initial conditions:

$$C_{A0} = C_{B0} = 0.1 \text{ N}$$

$$F_{A0} = F_{B0} = 80 \text{ ml/min.}$$

At time = 0:

Upset  $C_{A0}$  to 0.05Nand  $F_{B0}$  to 100 ml/min.

A: NaOH      B: Etac.

Temperature = 80°F

Time <u>Min.</u>	<u><math>C_{A0}</math> (g-moles/liter)</u>			
	<u>Run 1</u> <u>0 rpm</u>	<u>Run 2</u> <u>0 rpm</u>	<u>Run 3</u> <u>205 rpm</u>	<u>Run 4</u> <u>300 rpm</u>
0	0.01850	0.01900	0.02030	0.02080
1	0.01798	0.01861	0.01927	0.02042
2	0.01525	0.01572	0.01622	0.01708
3	0.01337	0.01297	0.01372	0.01441
4	0.01135	0.01111	0.01154	0.01265
5	0.01012	0.00988	0.01048	0.01120
6	0.00905	0.00920	0.00958	0.01019
7	0.00802	0.00861	0.00888	0.00950
8	0.00730	0.00771	0.00823	0.00874
9	0.00661	0.00730	0.00794	0.00821
10	0.00630	0.00670	0.00737	0.00798
11	0.00605	0.00648	0.00709	0.00741
12	0.00601	0.00624	0.00683	0.00722
13	0.00582	0.00615	0.00669	0.00703
14	0.00580	0.00601	0.00660	0.00690
15	0.00580	0.00601	0.00660	0.00690

C<sub>Ao</sub> (g-moles/liter)

Time	Run 5	Run 6	Run 7	Run 8	Run 9
<u>min.</u>	<u>433 rpm</u>	<u>582 rpm</u>	<u>890 rpm</u>	<u>1050 rpm</u>	<u>1550 rpm</u>
0	0.02140	0.02170	0.02202	0.02200	0.02200
1	0.02080	0.02041	0.02000	0.01931	0.01936
2	0.01698	0.01732	0.01741	0.01700	0.01698
3	0.01537	0.01542	0.01521	0.01523	0.01521
4	0.01292	0.01342	0.01375	0.01372	0.01377
5	0.01150	0.01221	0.01220	0.01221	0.01221
6	0.01052	0.01100	0.01111	0.01119	0.01120
7	0.00978	0.01019	0.01030	0.01031	0.01023
8	0.00902	0.00950	0.00951	0.00971	0.00953
9	0.00854	0.00908	0.00894	0.00924	0.00893
10	0.00793	0.00854	0.00873	0.00891	0.00861
11	0.00756	0.00811	0.00810	0.00850	0.00817
12	0.00731	0.00779	0.00778	0.00821	0.00789
13	0.00709	0.00745	0.00767	0.00801	0.00783
14	0.00706	0.00725	0.00749	0.00791	0.00749
15	0.00706	0.00720	0.00720	0.00758	0.00726
16	0.00706	0.00720	0.00720	0.00742	0.00720
17	0.00706	0.00720	0.00720	0.00720	0.00720
18	0.00706	0.00720	0.00720	0.00720	0.00720

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