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THE START-UP
OF
AN IMPERFECTLY MIXED
CONTINUOUS FLOW STIRRED TANK REACTOR

By
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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 Master of Science.

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ABSTRACT

Experimental investigation is made of the effect of imperfect mixing on the chemical conversion in a continuous stirred tank reactor. The performance of the reactor using a second order reaction is compared to that of a perfectly stirred tank, that of a plug flow reactor, that of a completely segregated model, and that of a mixed model obtained from tracer runs.

The inadequacy of residence time distributions to describe a non-linear reaction system is experimentally shown. Micromixing effects are discussed. The trend of higher conversion from complete segregation to maximum mixedness as influenced by the level of mixing is shown. The use of complete segregation to give a conversion limit is experimentally shown to be incorrect.

ACKNOWLEDGMENTS

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DEDICATION

To my parents, Franklin and Zoila, and to my brothers,
Guido and Fernando.

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INTRODUCTION

The level of mixing in a continuous flow reactor is a determining factor for the chemical conversion in the reactor. To incorporate mixing into the mathematical model of a reactor, it is necessary to have a model of the mixing process itself. Perfectly stirred tanks and plug flow reactors are the two ideal models of mixing, and their incorporation in reactor performance is of no problem. But the large number of cases where non-ideal mixing exists poses difficult and serious challenges.

In accounting for deviations from ideal flow it is desirable to have a simple model that will behave mathematically the same as the actual reactor. The use of residence time distributions to find such a model has been increasingly popular. For linear processes, where the rate of reaction is of first order, only the matching of residence time distributions of the model and the reactor is needed. This information obtained from residence time distributions is termed macromixing.

For non-linear processes, not only macromixing but also micromixing is necessary to describe the system. Micromixing fully explains the molecular scale behavior of the elements throughout passage of the reactor. Complex

kinetics, heating effects, and speed of reaction play important roles and are included in micromixing. In order to provide some information about micromixing which affects conversions in a reactor, complete segregation and maximum mixedness have been proposed to give limits for the conversion. Maximum mixedness is complete mixing to the molecular scale, while complete segregation is clustering of molecules in batchlike fashion with no interaction between clusters. Both of the above terms may be coupled with the residence time distribution of the system to give more exact limits. But the mathematical complications in finding a manageable function for the residence time distribution and in solving for the values of the limits with said function make the extension of questionable practicality.

Segregation is only meant to give some information about micromixing; the degree of segregation being defined as the variances of ages of all the molecules over the variances of ages of the clusters involved. Also, the fact that no variations in conversion have been found for premixed and separate reactant feed questions the applicability of complete segregation limits.

Therefore knowledge of micromixing must be coupled with residence time distributions for modeling of nonlinear reactions, although only very general concepts

describing micromixing are available. Experimental investigation should thus play an important role in the evaluation of micromixing effects on the use of models for non-linear systems. This project aims at providing experimental work for such an evaluation.

LITERATURE SURVEY

The literature on the effect of mixing or agitation on chemical reactor performance had its first significant contribution in MacMullin and Weber's paper⁽¹⁾ in 1935. Their paper dealt with the residence time distribution of a flow system of a number of tanks in series, and with short circuiting in such a system. It was 1953 when Danckwerts' treatment of residence time distributions in continuous flow systems⁽²⁾ brought the topic into focus again, and signaled a continued interest that has brought a voluminous number of publications in the area.

The treatment of mixing of fluids in a vessel has been confined to the approach by models, and not to the more rigorous approach using fundamental hydrodynamics. This is due to the great mathematical difficulties encountered, which make the latter approach impractical.

The mathematical representation by models of fluid mixing in a flow system must consider macromixing - retention time characteristics of the fluid on the system, and micromixing - individual particle behavior, such as probability of two particles coming together. Macromixing is fully defined by the residence time distribution, which as noted previously was the initial area of interest^(1,2).

Micromixing has been defined⁽³⁾ in terms of residence time distribution characteristics as the transition of groups of molecules entering the reactor with equal ages but different life expectations to groups leaving the reactor having equal life expectations but different ages. Danckwerts⁽⁴⁾ first recognized the influence of micromixing and introduced segregation as a measure of micromixing. Two other parameters have since been introduced as a measure of micromixing^(3,5). Danckwerts⁽⁴⁾ also defined complete segregation as a complementing and opposing state to complete mixing down to molecular scale. Greenhalgh and others⁽⁶⁾ independently arrived at the same facts as Danckwerts, called the new state zero intermixing, but their work has not attracted much attention. Zwietering⁽⁷⁾ tied the micromixing-to-molecular-scale state to an arbitrary residence time distribution, and defined it as maximum mixedness.

The numerous models presented to describe non-ideal mixing in a stirred tank may be divided in the following four categories. First, models based on the two ideal reactors, plug flow and stirred tank. The plug flow with axial diffusion (dispersed plug)⁽⁸⁾, the tubular with recycle^(9,10), the tanks in series⁽⁸⁾, the tanks in series with recycle⁽¹¹⁾, and the tubular-stirred tank with recycle⁽¹²⁾ are some of the models in this first category.

Also included in the category are the combination or mixed models^(13,14,15,16). The models in this group are made up of simple regions (plug flow, perfect mixing, dispersed plug flow, or dead space) interconnected by various flow streams (bypass, recycle, or crossflow).

Second, models that attach much importance to the effect of the stirrer on its immediate surroundings, and that either separate the reactor in two flow regions, or use loops to account for recirculation of flow. Van de Vusse's model⁽¹⁷⁾ is the most prominent one in this category, and other models closely resembling it have been proposed^(18,19,20). The significance of the two parameters in these models has been experimentally shown^(21,22,23). Another interesting model in this category is that presented by Manning and others⁽²⁴⁾, while the rest of the models are more restricted^(25,26,27).

Third, models that heavily rely on probability to describe the performance of the system^(28,29,30).

Finally, generalized models that attempt to describe the mixing process as fully as possible^(3, 31,32). One general model of unique importance is that of Ng and Rippin⁽³³⁾, consisting of complete segregation and maximum mixedness environments, and a transfer parameter between environments that determines the extent of micromixing (or

conversion) in the reactor. This model allows for changes in degree of segregation independent of residence time distribution changes.

The application of models in the explanation of non-ideal systems has been suggested⁽³⁴⁾ as follows: use of dispersed plug model, which may be considered the equivalent of a number of stirred tanks in series⁽⁸⁾ for small deviations from ideality, and use of mixed models for larger deviations. The actual application of models⁽³⁵⁾ has two sources: in the design of a reactor, where a model is made to fit expected flow condition of the physical system and predicts the performance of such system; and in the testing or analysis of an existing reactor, where obtained residence time distribution curves determine the model that describes the reactor. For linear systems the application of models is sound, but when reactions of order higher than one are present an uncertainty factor is introduced due to micromixing effects. Danckwerts⁽⁴⁾, Zwietering⁽⁷⁾, Weinstein and Adler⁽³⁾, Adler and others⁽³⁶⁾, and Rippin⁽³⁷⁾ discuss this characteristic in depth, the latter concluding that reactors having same residence time distributions and same degree of segregation may nevertheless produce different degrees of conversion, due to the fact that degree of segregation does not represent fully

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concentration history of the elements in the reactor.

Novosad and Thyn⁽³⁸⁾ developed charts that give the conversion limits for backmix reactors in series.

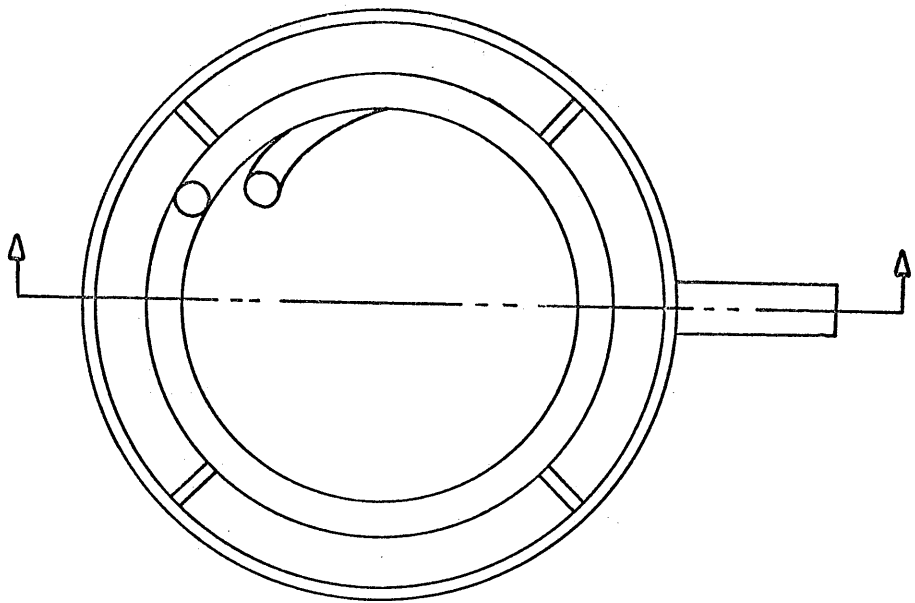
Experimental work related to the use of models to describe flow reactors has been somewhat limited. Cholette and Cloutier⁽¹³⁾, and Bartock and others⁽³⁹⁾ presented work with linear systems. LaRosa and Manning⁽⁴⁰⁾ performed exploratory work in a system with a second order reaction, found that experimentally measured rates of reaction decrease with increasing level of mixing, and attempted to correlate the degree of segregation in the system with rpm. Worrell and Eagleton⁽⁴¹⁾ also noted that conversion values in a similar system increase from the region of maximum mixedness to that of complete segregation with decreasing rpm. Ng and Rippin⁽³³⁾ performed preliminary work in this area, while Adler and others⁽³⁶⁾ obtained supporting results to the use of conversion limits for a tubular reactor represented by a mixed model. The need for further experimental investigation, especially in the nonlinear cases, has been widely recognized^(33,36,40).

EXPERIMENTAL WORK

The experimental work may be described in four sections: apparatus, chemicals, procedure for tracer runs, and procedure for reaction runs.

Apparatus

The reactor, the main piece of apparatus and shown in Figure 1, consisted of a 925-ml stainless-steel, cylindrical vessel mounted with baffles and coils. The four baffles were equally spaced and their width was one-tenth of the vessel diameter. The $\frac{1}{2}$ x $\frac{1}{4}$ -in.-O.D, copper-tubing coils made five full turns inside the reactor, and were soldered to the baffles. The coils exited the reactor through the top. The impeller was a 1-in.-diameter, stainless steel, three-blade propeller (Sargent S-76701 A), centered in the reactor, which was driven by a Universal motor implemented with a solid-state speed control and a permanent magnet d-c tachometer. The stirring motor had a maximum speed of 6000 rpm. A U4-1 Design Corporation speed reducer of 4.96-to-1, input-to-output ratio was used for stirring speeds below 1000 rpm to minimize variations in the stirring rate during each run. The reactor-impeller system was designed, following Chapman and Holland⁽⁴²⁾, to avoid vortex formation and to provide vigorous mixing below 3500 rpm.



TOP VIEW

ALIGNED SECTION

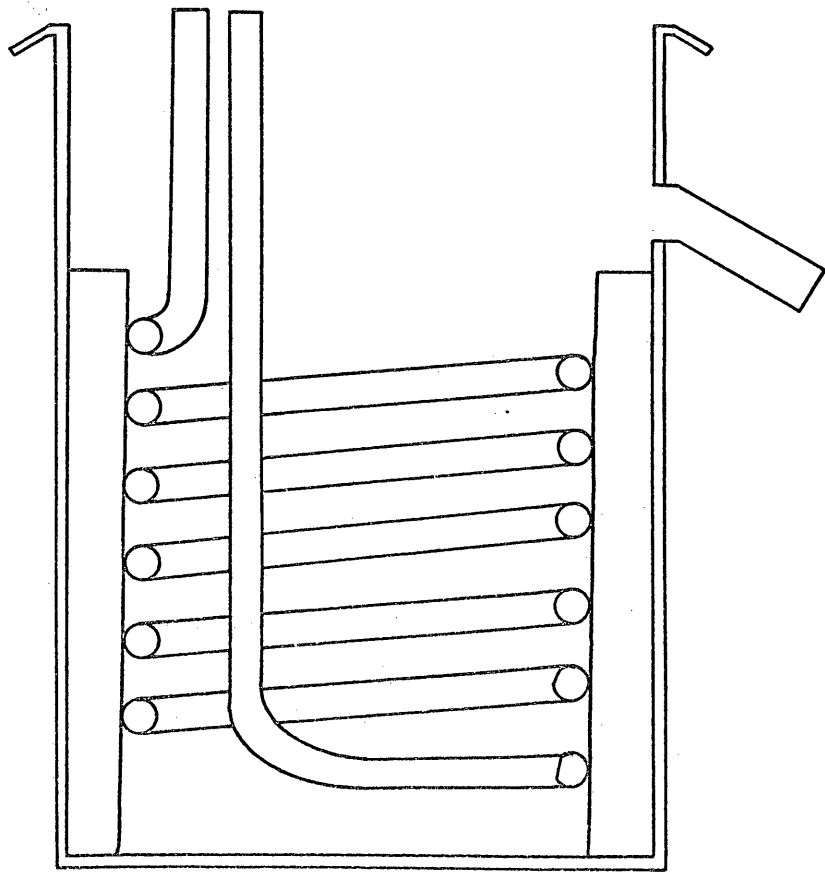


FIGURE 1 - REACTOR SYSTEM

DIMENSIONS FOR FIGURE 1.

Vessel OD	4 1/8 in.
Vessel ID	4 in.
Vessel height	5 3/4 in.
Outlet height	4 1/2 in.
Outlet size	3/8 in. st. st. pipe
Outlet pipe length	1 in.
Angle of outlet	30°
Baffle width	13/32 in.
Baffle height	4 in.
Coils	1/4 in. copper tubing
Coils bottom level	1/2 in. from vessel bottom
Coils top level	3 3/4 in. from vessel bottom
Impeller	three-blade propeller
Impeller diameter	1 in.
Impeller position	centered
Impeller height	1 in. from vessel bottom

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The flow system for the entire apparatus is shown in Figure 2. Reagents were fed to the reactor by an air-pressure, displacement method. The reagents were stored in 5-gal. bottles, with $\frac{1}{4}$ -in. copper-tubing connections to the air-feed line in one end, and to a rotameter in the other end. The air pressure to the storage bottles was controlled by a needle valve and then by a pressure regulator. A pressure gage was used to check the pressure in the storage bottles. The rotameters used to measure reagent flow were Hicksville rotameters of 20 to 200-cc per min range. The copper-tubing feed lines following the rotameters were then submerged in a constant temperature bath, so that the temperature of reagents going into the reactor would be constant. Polyflo tubing was used at the end of the feed lines to provide flexibility in the connections to the reactor.

The temperature in the reactor was measured by a copper-constantan thermocouple connected to a Hewlett-Packard, Model 7100B, strip-chart recorder. To continuously monitor the concentration of products leaving the reactor a Leeds and Northrup pH meter and a Sargent miniature combination electrode were used. The output of the pH meter was connected to the recorder mentioned previously.

An automatic titrator was used in all titrations.

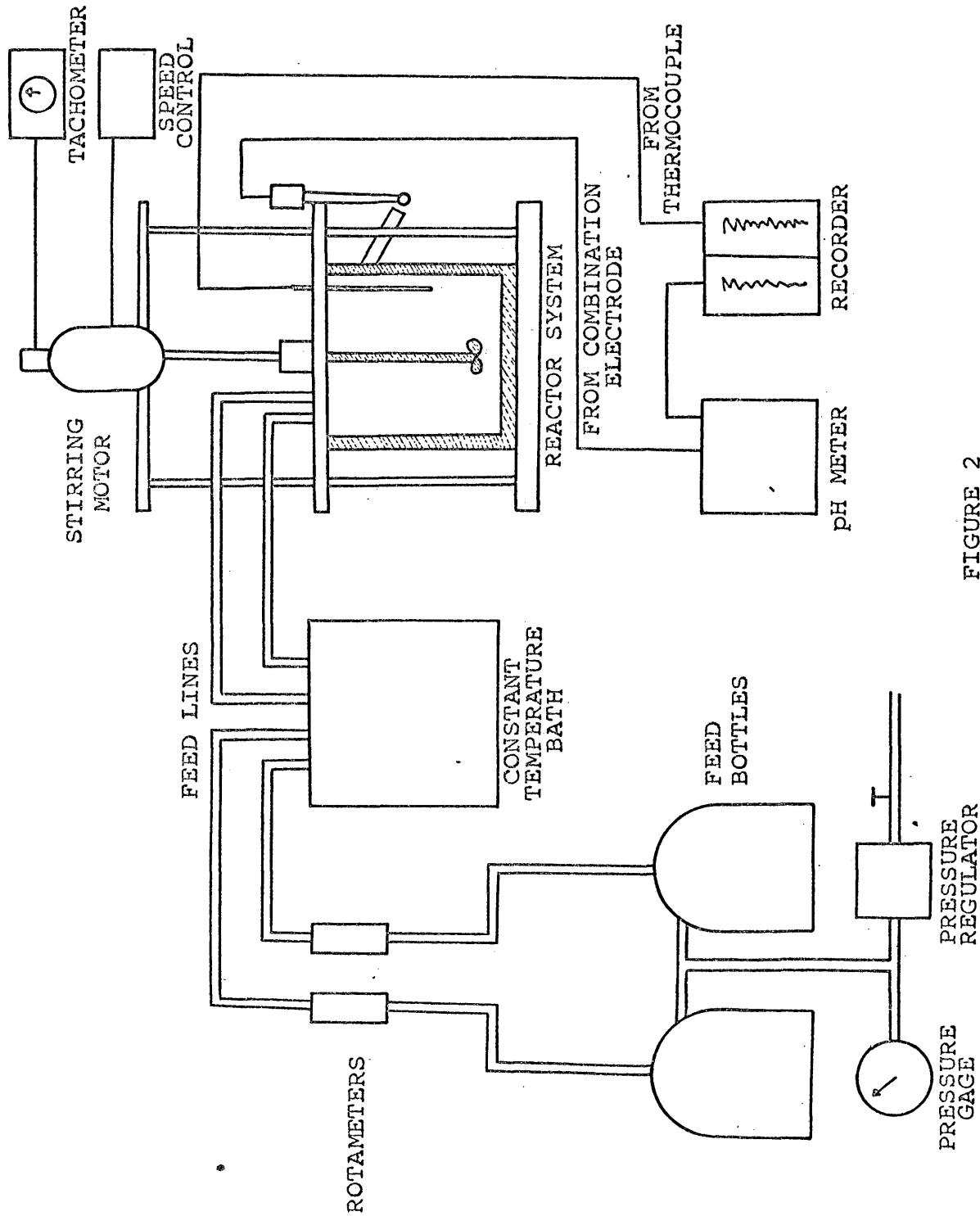


FIGURE 2
REACTION FLOW SYSTEM

Chemicals

Approximately 0.1 N sodium hydroxide and distilled water were used in the tracer runs. The choice of these solutions was based on their very close proximity in density and viscosity to reactants and products in the reaction runs. (See Appendix I)

Approximately 0.1 N ethyl acetate solution, made from Mallinckrodt analytical reagent grade ethyl acetate, and 0.1 N sodium hydroxide solution were the reagent solutions. As the ethyl acetate hydrolyzed even in the absence of base, the solution had to be used shortly after preparation.

Standard hydrochloric acid and standard sodium hydroxide were used in analysis and titrations.

Procedure for tracer runs

The approach used to obtain a model of the reactor from its residence time distribution was that presented by Cholette and Cloutier⁽¹³⁾. The method involved a negative step change in the feed concentration to provide an F-curve.

The reactor system was started at a given rpm and at 200-ml-per-min flow of sodium hydroxide solution. After a few minutes, when all flow patterns were assumed to be fully developed the feed was changed to an equal flow of distilled water. The pH of the outlet stream was recorded, and several samples were also taken at regular time inter-

vals. These samples were later titrated to give a check on the pH results for sodium hydroxide concentration of the outlet stream as a function of elapsed time.

Procedure for Reaction Runs

To start the reaction runs, initially the reactor was full of equimolar amounts of reactant solutions, and continuous feed of the reactants was begun at the time the reaction commenced. This path was chosen to afford a continuous decrease in reactant concentration throughout the course of the process, and to allow for constant stirring and recording of solution pH from the beginning of the reaction.

The reactor was first half-filled with sodium hydroxide solution. An equal volume of ethyl acetate solution was then dumped into the reactor. This dumping of solution took approximately six seconds. During this period the reactant feed lines were connected to the reactor, each reactant flowing at a rate of 100 ml per min and of same concentration as that of the solutions used to fill up the reactor. Also during this interval, stirring was initiated and set at the desired level. For stirring rates below 200 rpm, the stirring was started after the reactor was only half-full. In these cases the stirring did not splash the solution when the reactor was partially full, which

would have been the result at higher rpm.

Temperature of the solution was measured inside the reactor. Negligible heating effects occurred at the low reactant concentrations.

The pH of the solution was measured at the outlet of the reactor by allowing the stream to flow freely over the tip of the miniature electrode. A few samples were taken from the outlet stream during the approach to steady state in the reactor. These samples were analyzed for sodium hydroxide concentration; one sample was used to check the difference between sodium hydroxide and ethyl acetate concentrations. The time required for approach to steady state was about 6-to-9 minutes. The system was allowed to run for approximately another 15-to-20 minutes, during which period samples were taken to check on the steady state values. Two-to-three samples were obtained during the approach to steady state, and about five samples at the steady state. These samples were used as a check on the recorded pH values.

The analysis of reaction samples consisted of the accepted method of quenching the reaction, and titration with standard acid to give the sodium hydroxide concentration. In order to obtain the ethyl acetate concentration, the sample was added to a given amount of excess sodium

hydroxide, refluxed to complete the reaction, and titrated to give the final excess sodium hydroxide. The ethyl acetate concentration could be found using the previously obtained sodium hydroxide concentration, but the main interest was to check the relative concentration differences.

The reactants were analyzed using slight modifications of the method just described. The ethyl acetate feed solution was analyzed immediately before each run to avoid concentration errors caused by the uncatalyzed hydrolysis.

RESULTS AND INTERPRETATION

This section may be divided into results and interpretation of tracer runs, and results and interpretation of reaction runs.

Tracer Runs

Data obtained from the tracer runs are tabulated in Appendix II. These data were plotted according to Cholette and Cloutier⁽¹³⁾ to obtain a mixed model that fits the tracer run information (Figures 3 to 6).

The plots show that there are two regions where the general characteristics of the curves differ markedly. For runs below 100 rpm, the curves show very large and sudden declines in the outlet concentration, which later on taper off. For runs at 200 rpm or above, the decline in reactor concentration was very close to the exponential form of a perfect mixer. In the range of 100-to-150 rpm, there were marked variations in the shape of decline curves for a given rpm, and it was concluded that this range was a transition zone in flow behavior between higher and lower stirring rates.

The shape of the curves in Figure 3 indicated that a fairly complex mixed model would be necessary to match the reactor performance at mixing rates below 150 rpm as

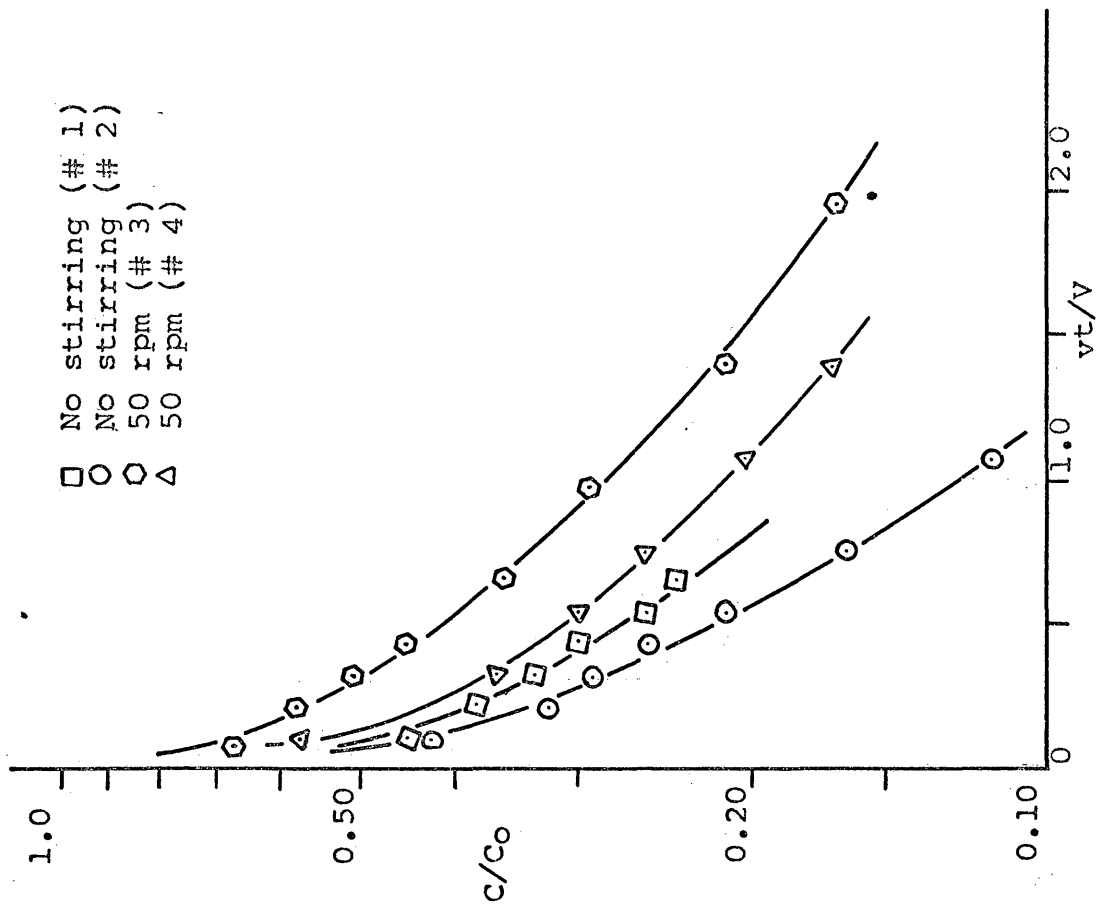


FIGURE 3
TRACER RESULTS
0 & 50 rpm

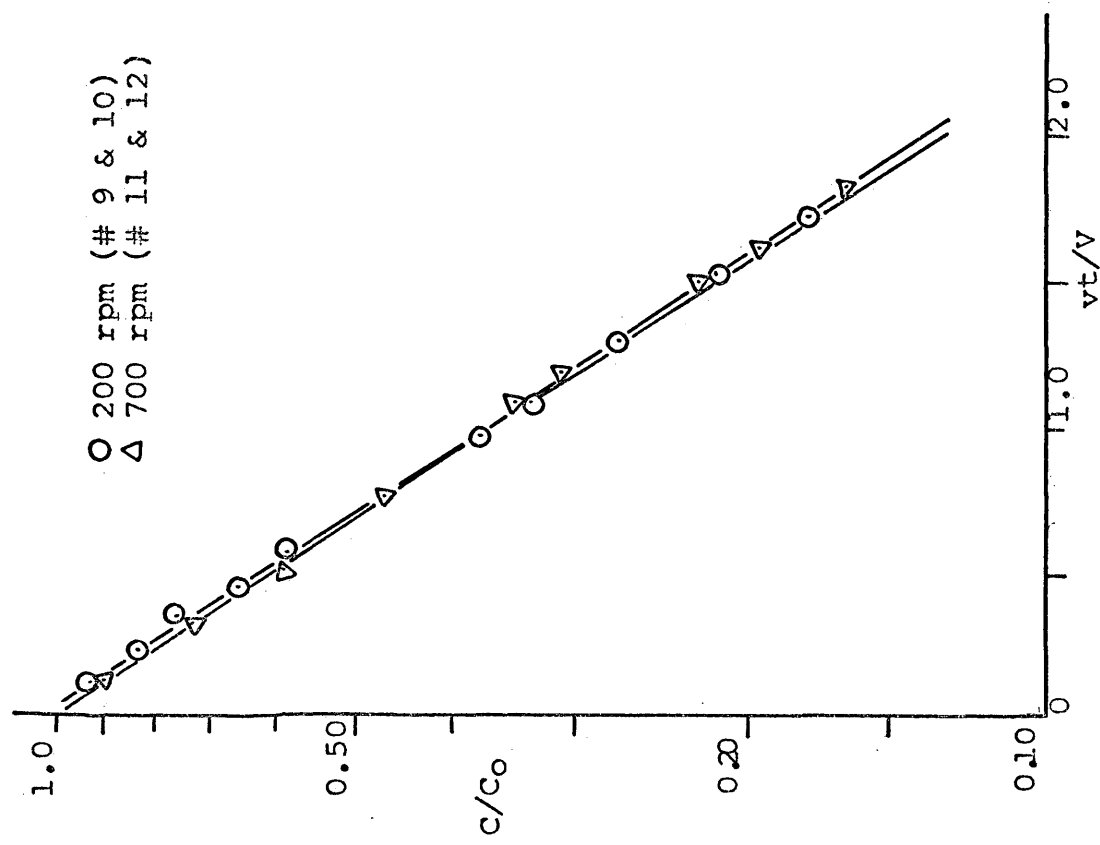


FIGURE 4
TRACER RESULTS
200 & 700 rpm

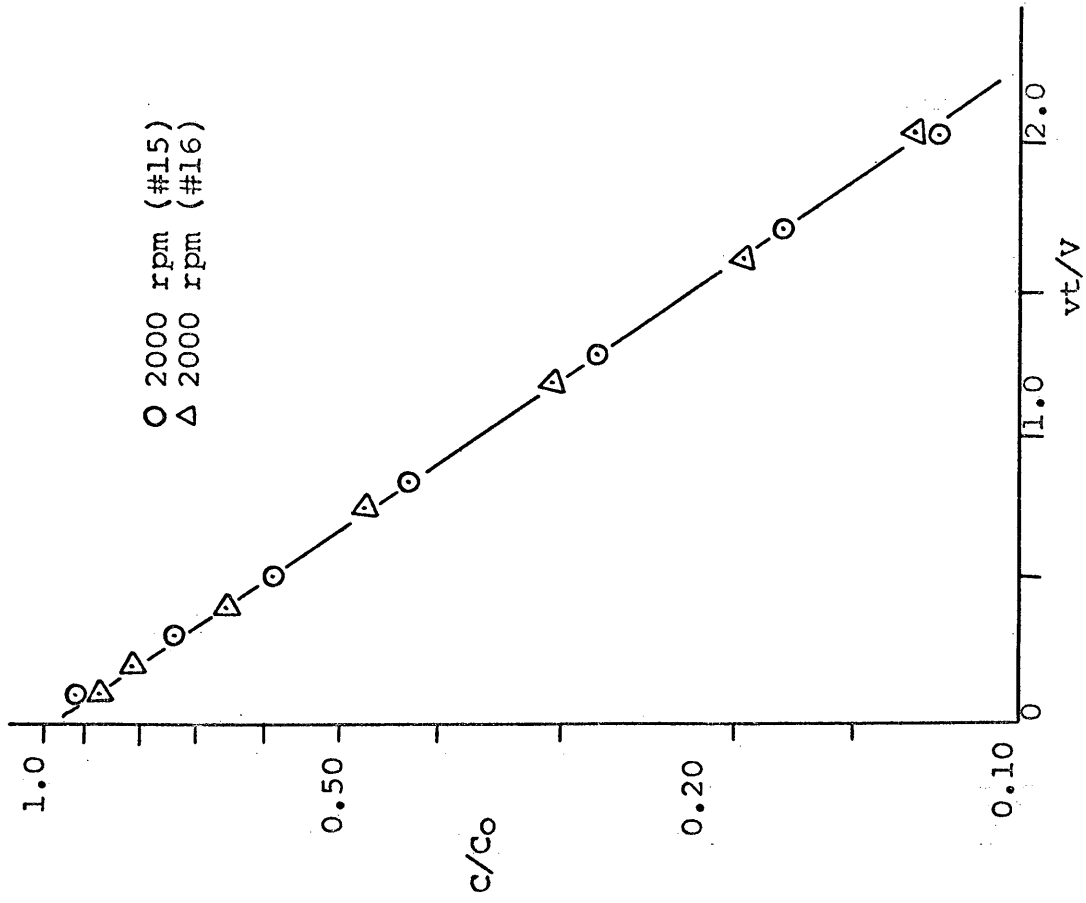


FIGURE 5
TRACER RESULTS
2000 rpm

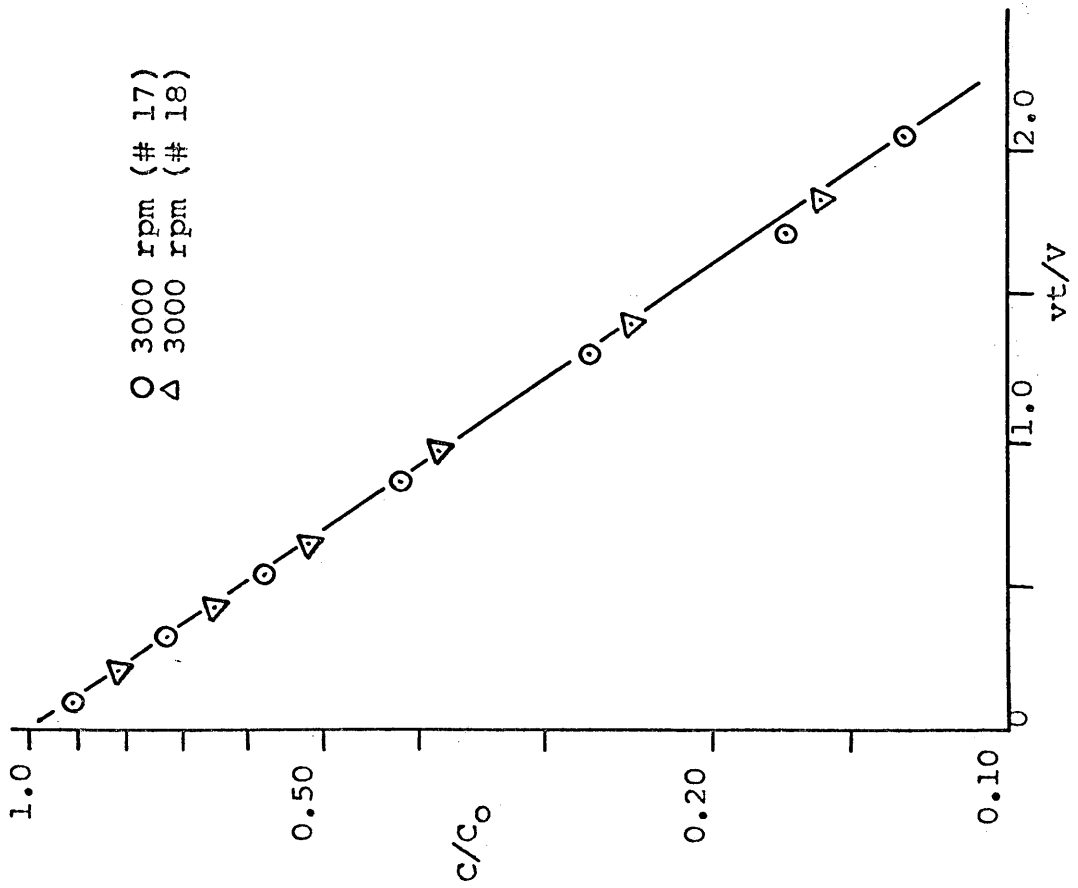


FIGURE 6
TRACER RESULTS
3000 rpm

evaluated by the tracer runs. Trial and error testing of various mixed models with an analog computer would be essential if very close matching of experimental and mixed model performance was desired. Since there were some variations in curves obtained at a constant rpm, obtaining a very close matching of curves was ruled out. Mixed models that approximately correspond to reactor performance were then obtained, as outlined in Appendix III. Figure 7 shows the general model that fitted low-stirring-speed curves (below 100 rpm), and Table I gives the model parameters for the different stirring speeds.

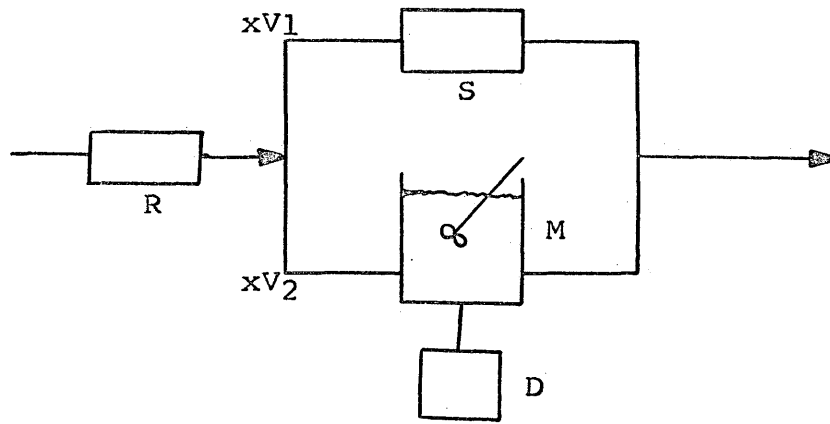
TABLE I

Low-Mixing-Range Model Parameters

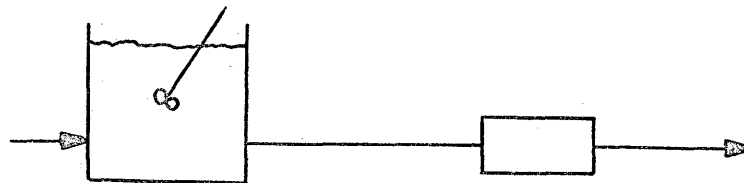
RPM	R	S	M	D	xV_1	xV_2
0	.025	.010	.35	.615	.50	.50
50	.025	.010	.875	.090	.30	.70
100	.025	.010	.965	--	.25	.75

R, S, M, and D are fractions of reactor volume.

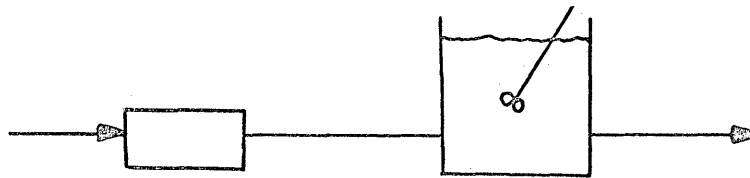
xV_1 and xV_2 are fractions of total-flow through reactor.



COMBINATION MODEL LOW rpm RANGE



STIRRED-TUBULAR MODEL (S-T)



TUBULAR-STIRRED MODEL (T-S)

FIGURE 7
MIXED MODELS FROM TRACER RESULTS

The general model obtained for higher stirring level consisted of partial perfect mixing coupled with partial plug flow. As the two different configurations of this general model could affect its performance both alternatives had to be considered for further work. Thus the stirred tank followed by plug flow (S-T model) and the plug flow followed by a stirred tank (T-S model) were the resulting models, as shown in Figure 7. The parameters for these models (volume fractions) were obtained from the slope and intercept of the lines in Figures 4, 5, and 6. The least squares fit method was used to find the slope and intercept of the lines. Table II shows the resulting parameters for all the rpm values in the higher range.

TABLE II

<u>RPM</u>	<u>T-S Model</u>		<u>S-T Model</u>	
	<u>T</u>	<u>S</u>	<u>S</u>	<u>T</u>
200	.038	.962	.962	.038
400	.021	.979	.979	.021
700	.010	.990	.990	.010
1250	.009	.991	.991	.009
2000	.008	.992	.992	.008
3000	.009	.991	.991	.009

T and S are fractions of reactor volume.

The standard deviation in the above values is of the order of .0005.

Reaction Runs

Curves for the approach to steady state for the reaction runs are shown in Figures 8 through 15. These curves were obtained both from concentration values of the samples taken during reaction, and from correlation of pH values based on the sample points. A correlation was drawn for every run, and it was necessary to do so as the outlet stream was flowing freely over the tip of the combination electrode, and the exact position of the electrode relative to the stream was a determining factor in the values recorded. Once the reaction was started, care had to be taken that the combination electrode position was not altered. Recordings of the pH values were almost without exception very smooth.

Along with the experimental curves, approach to steady state by a perfectly stirred tank reactor and by a plug flow reactor are shown in Figures 8 through 15. In the low rpm range the approach to steady state by the appropriate combined model obtained from the tracer runs is also plotted. For the high rpm range only the steady state value given by the combined model is added. This value corresponds to the T-S model for the mixing level, since the T-S model gave a greater variation from the perfectly stirred tank value than the S-T model. The approach to

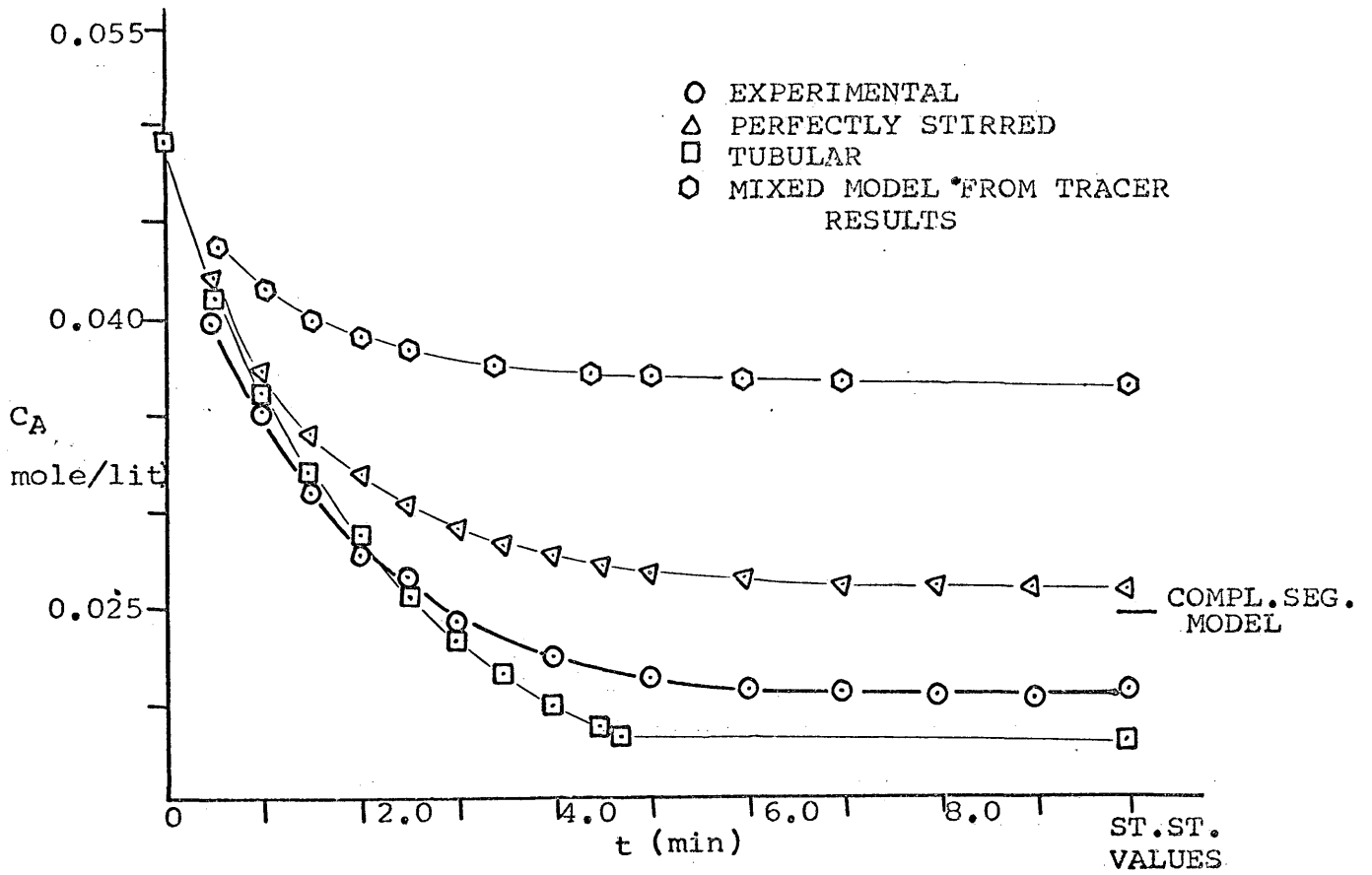


FIGURE 8
 RUN # 3 - NO STIRRING

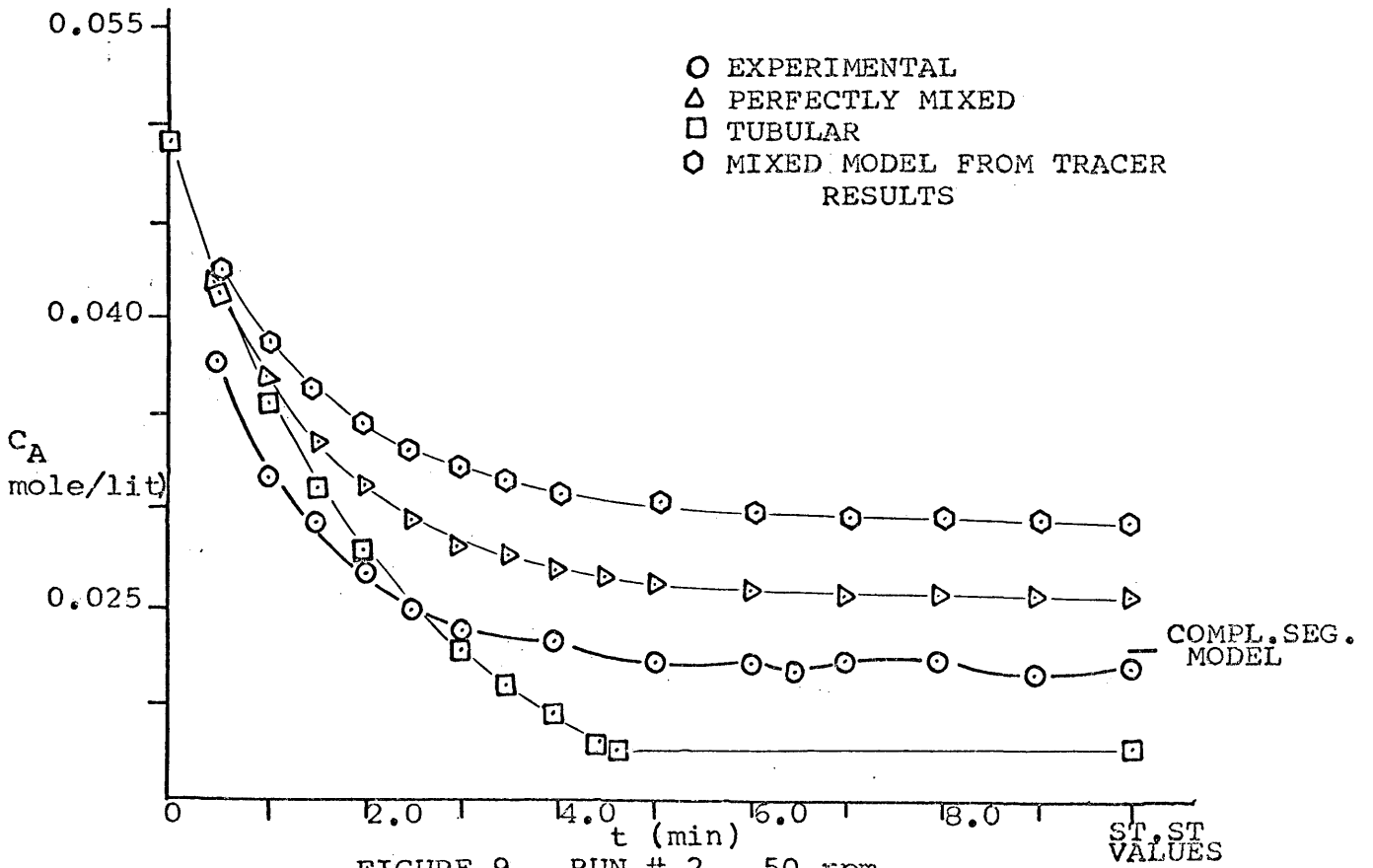


FIGURE 9 RUN # 2 - 50 rpm

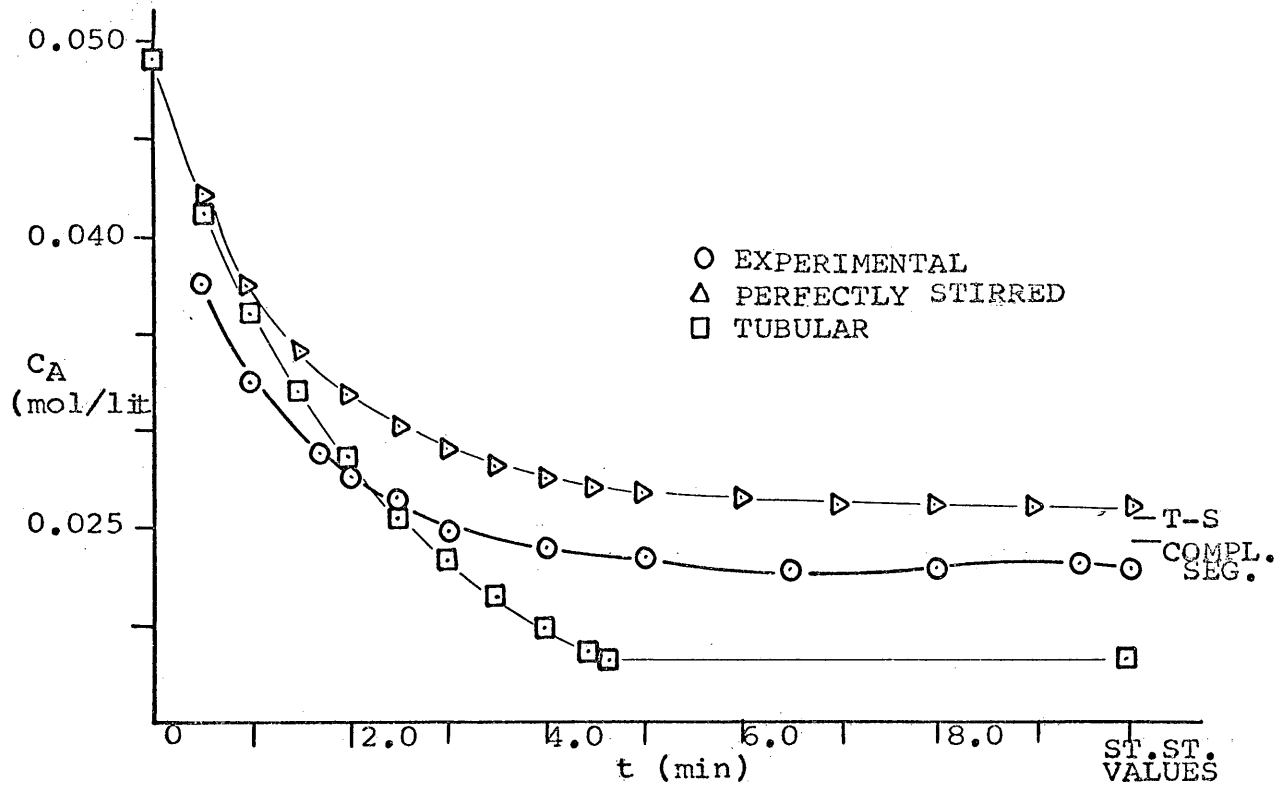


FIGURE 10
 RUN # 5 - 200 rpm

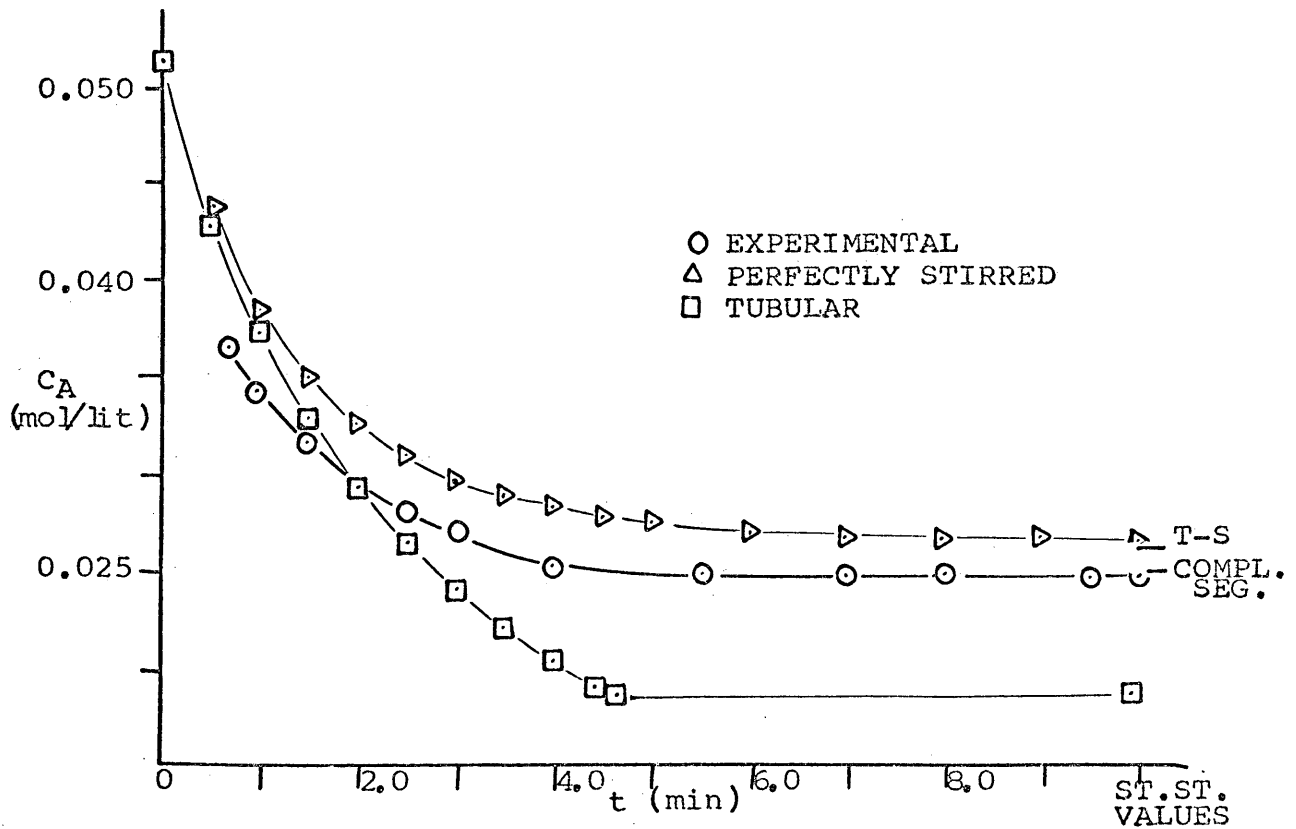


FIGURE 11 RUN # 7 - 400 rpm

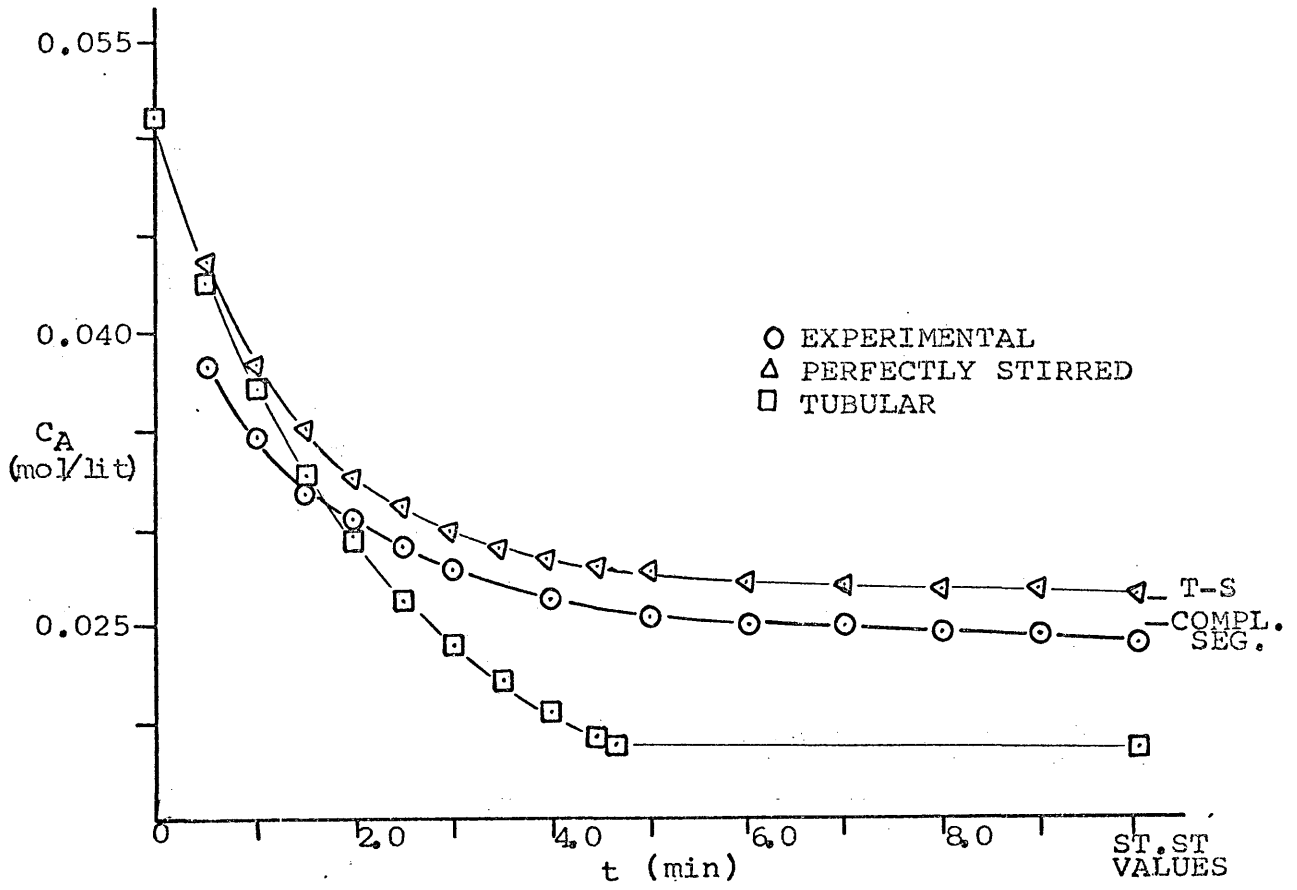


FIGURE 12
 RUN # 9 - 700 rpm

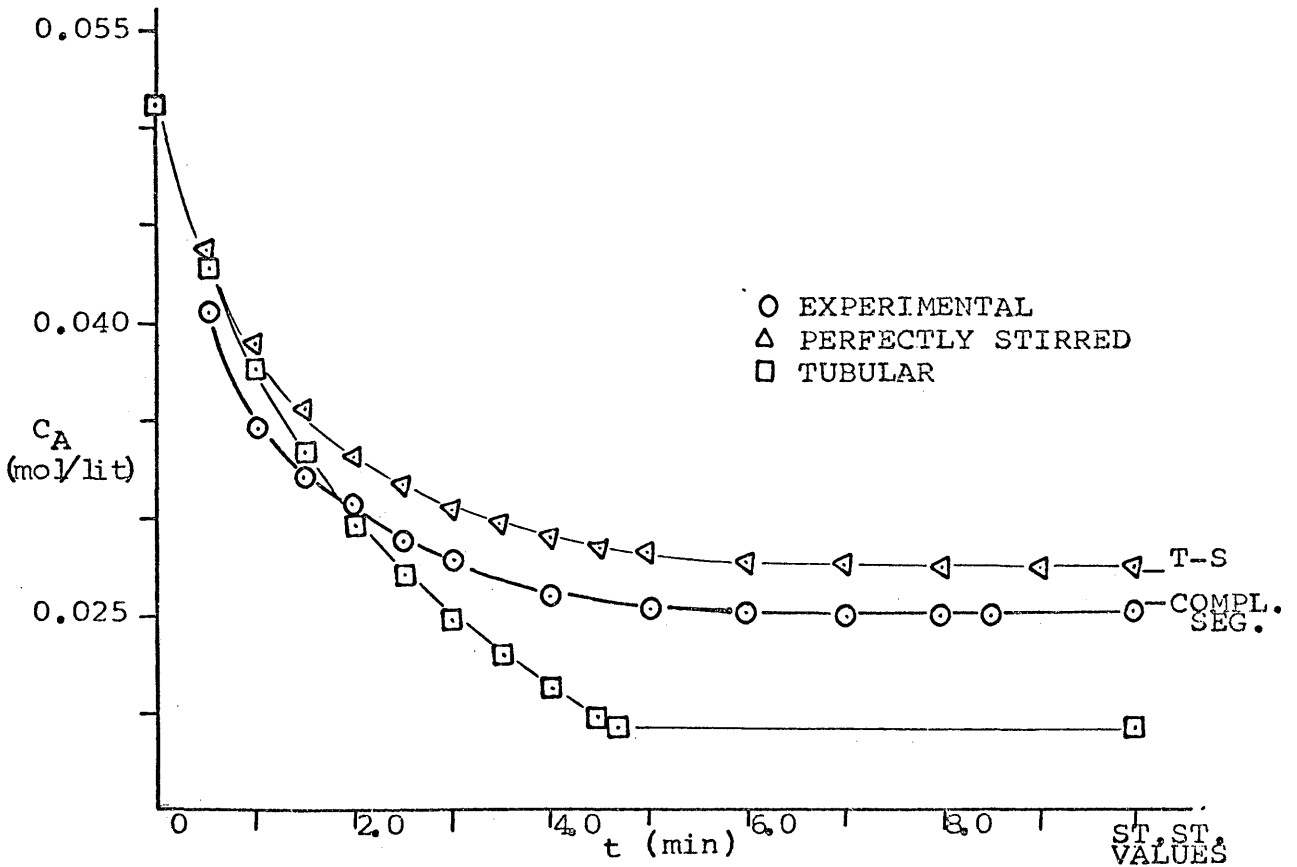


FIGURE 13 RUN # 16 - 1250 rpm

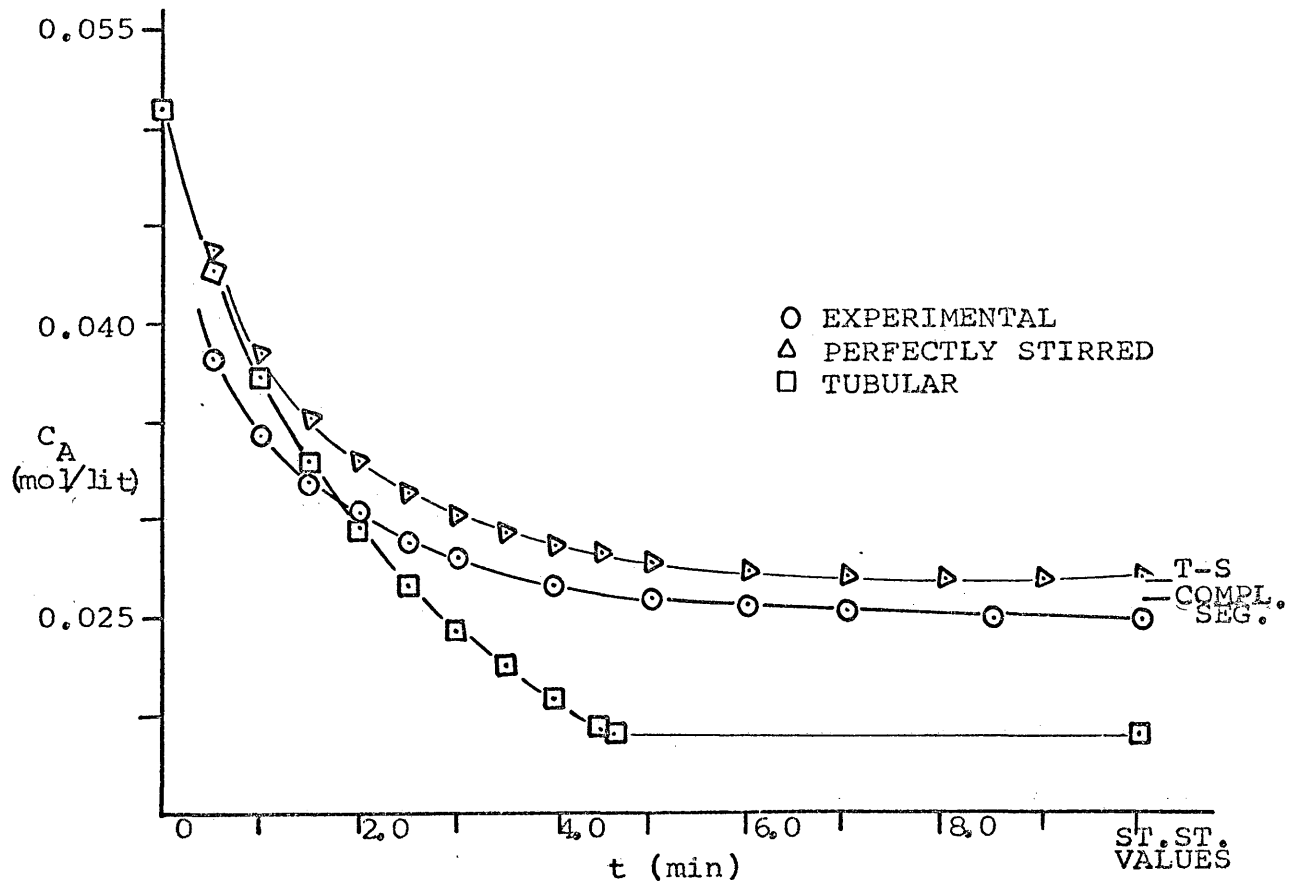


FIGURE 14
 RUN # 16 - 2000 rpm

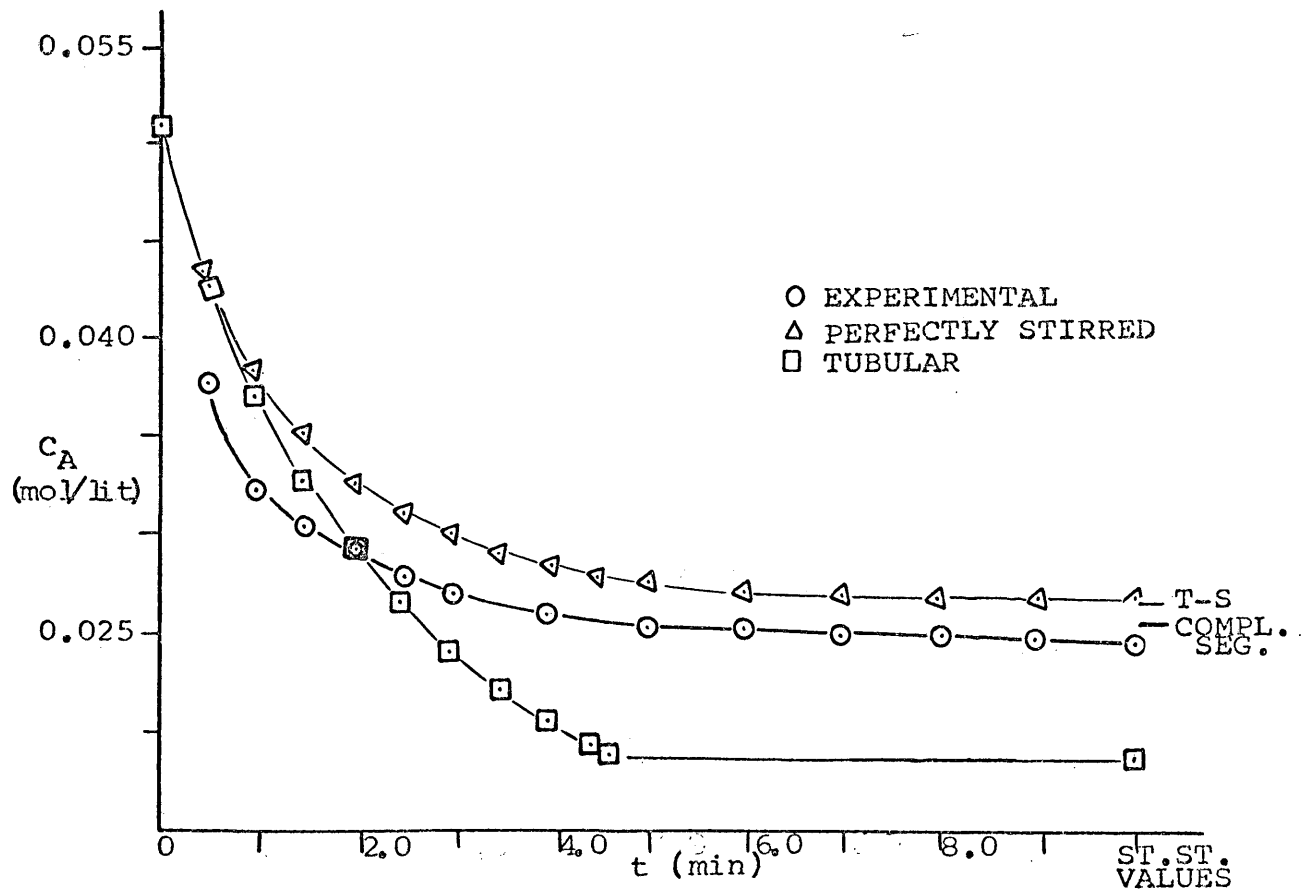


FIGURE 15 RUN # 10 - 3000 rpm

the steady state was not plotted because the variation from the perfectly mixed model was small. Also in these figures, the steady state value for a completely segregated model is shown. These values were obtained from correlations given by Novosad and Thyn⁽³⁸⁾, as shown in Appendix V. The accuracy of these values of only three significant figures was set by these graphical correlations.

A comparison of the experimental steady state values with both perfect stirred tank values and complete segregation values is given in Figure 16. Differences in steady state values are shown for both cases. Since initial concentration of reactants was not equal for all runs, fractional conversion would not give an exact basis for comparison and was not used.

Figure 16 shows that the sodium hydroxide concentration difference between a perfect mixer and the experimental values decreased with increasing rpm. There was marked decrease in the concentration differences up to about 200 rpm, after which the differences seemed to stay constant. Thus there were greater conversions at very low rpm values, where the tracer runs indicated distinct deviations in residence time distributions from that of a perfectly mixed model. A very initial response would be to find these facts illogical. Micromixing effects, and the con-

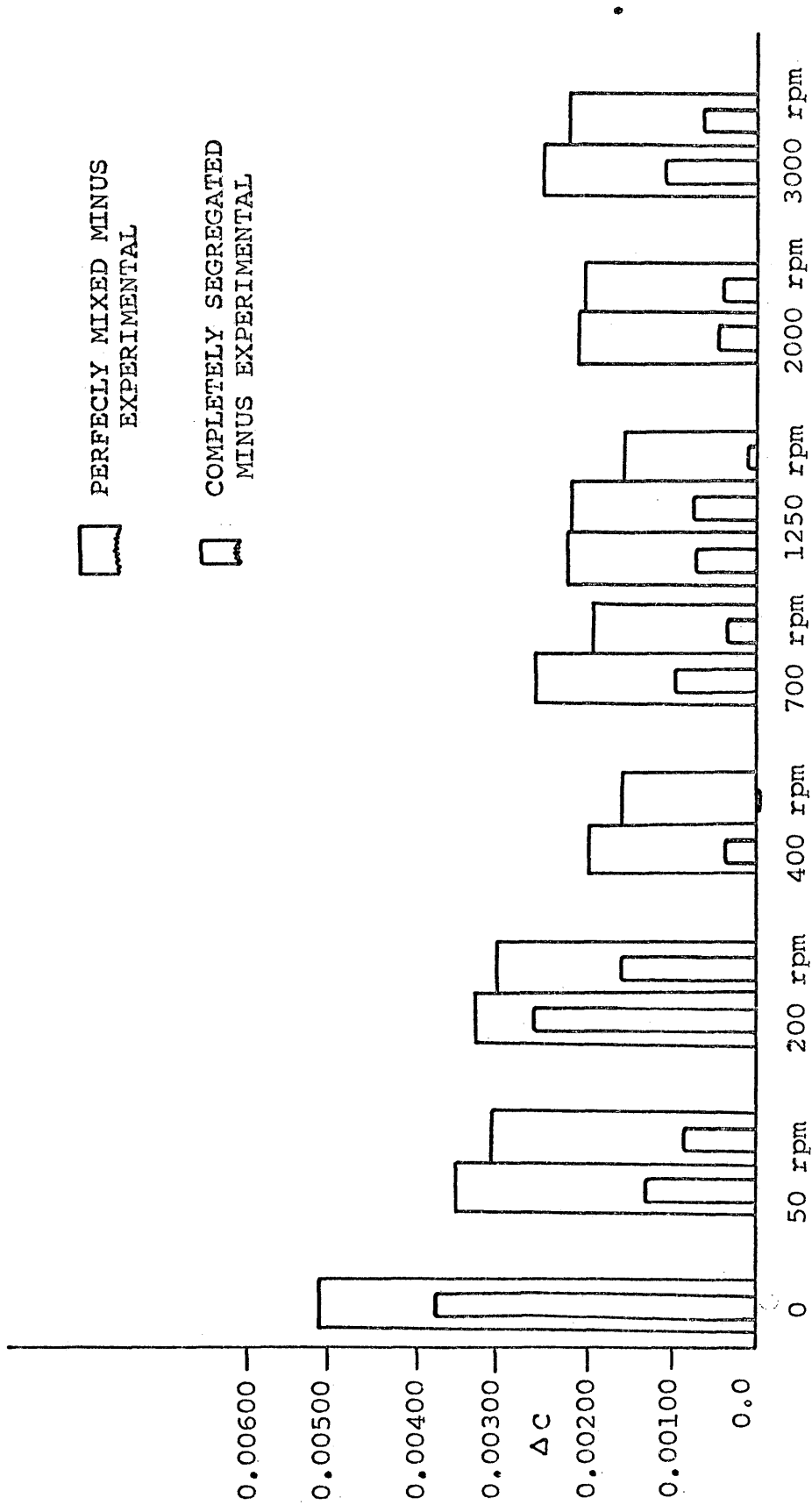


FIGURE 16
STEADY STATE CONCENTRATION DIFFERENCES

cept of segregation in particular, could however be used to discuss the results obtained.

Micromixing aims to describe the behavior of molecules during their passage through the reactor. As residence time distribution functions do not describe the measure of micromixing in the reactor, they cannot be used to predict the performance of a system where reaction requires collision of reacting species. This fact is shown very clearly in Figures 8 and 9. The deviations in the results given by models found by tracer studies from the experimental results are very large and in error of up to 28% conversion. (See Figures 8 and 9) The important characteristic of quick passage of flow through the reactor noticed in the tracer runs is the main reason for the low conversions predicted by the model. But the experimental results show that the transfer and use of that characteristic will not explain the behavior of the reaction system.

The concept of segregation partially describes micromixing by showing an aspect of the concentration properties of clusters of molecules. Complete segregation has been proposed as a condition of micromixing that gives a limit of conversion for a reacting system, based on the assumption that the batch-like behavior of the clusters signifies maximum reaction. The experimental results do not support

the use of complete segregation in the manner stated above. The experimental conversions were always higher than those predicted by complete segregation with only one exception where the difference was very small (0.1% conversion). The deviations between experimental and complete segregation values averaged about 1.5% conversion, while those between experimental and perfect mixer values averaged about 6% conversion.

The results thus show that although segregation provides some information about the micromixing in a system, its application must be coupled with a knowledge of the risks and uncertainties involved. Also it is shown that limits of conversion cannot be exactly determined by complete segregation conditions.

Other experimental work that supports these observations is that of Worrell and Eagleton⁽⁴¹⁾, who noticed a pattern in the conversions from complete segregation at lower mixing values to maximum mixedness at higher rates. But a large portion of the points from the above work that set the pattern falls below or above the limits of conversion. Thus the pattern is substantiated, but the validity of the limits may be questioned. Evidence for the pattern is also given in LaRosa and Manning's paper⁽⁴⁰⁾, but comparison with the limits was not possible. Preliminary

experiments by Ng and Rippin⁽³³⁾, consisting of two runs, also pointed out that conversion was higher for the lower-rpm run. But the conversion for the higher-rpm run was below that predicted by maximum mixedness. Adler and others⁽³⁶⁾ present experimental investigation of the conversion in a short tubular reactor which can be represented by a mixed model. In this case, the value for the experimental conversion did fall between limits of complete segregation and maximum mixedness for the mixed model.

Experimental work therefore proves the establishment of micromixing effects. For the reactions studied (second order, no competing or consecutive reaction, no heating effects, fast rate of reaction) these effects set a pattern of lower conversion with increasing rpm, pattern that agrees with the trend given by the limiting values of complete segregation as they are influenced by the residence time distributions⁽⁴¹⁾. But the limiting values themselves are not correct. This inaccuracy of complete segregation values should, however, not be astonishing, since segregation is based on variance of ages of elements in the system. Therefore segregation does not fully describe the concentration history of these elements, history which is required for the exact determination of the performance of the system.

CONCLUSIONS

The following conclusions may be drawn as a result of this investigation:

First, as is well accepted in the literature, residence time distribution functions cannot be used to predict reactor performance of a nonlinear system. This fact was confirmed very decisively in the experimental system used.

Second, the concept of segregation in a system is useful insofar as it provides some general characteristics of the micromixing.

Third, the complete segregation model does not correspond to the upper limit of conversion in a reactor, as shown by the experimental work.

Fourth, a great deal of derivation and definition of basic concepts that characterize micromixing is needed.

Finally, experimental work in the area is still rather limited. No work has been done in nonlinear systems with complex kinetics--such as side reactions or consecutive reactions--where very interesting complications seem to arise. Many other experimental areas are still untouched.

APPENDIX I

Density and Viscosity Comparison of Solutions

T = 25°C.

<u>Solution</u>	<u>Density, g/cm³</u>	<u>Viscosity, cp</u>
Water	.997044	.8956
0.1 N Sodium hydroxide	1.00252	.9119
Reaction products (.1 N NaOH + .1 N EtAc)	.99778	.9034

Water and Sodium hydroxide values from International Critical Tables. Products values determined by ASTM Kinematic Viscosity Test (D445).

APPENDIX IITRACER RUNS DATARun #1 - No stirring

<u>t</u>	<u>C</u>	<u>C/C₀</u>	<u>vt/V</u>
.50	.044691	.43872	.10810
1.00	.038623	.37915	.21622
1.50	.033802	.33182	.32432
2.00	.030314	.29758	.43243
2.50	.025748	.25276	.54054
3.00	.024103	.23661	.64865

Run #2 - No stirring

<u>t</u>	<u>C</u>	<u>C/C₀</u>	<u>vt/V</u>
.55	.042990	.42202	.11892
1.00	.032611	.32013	.21622
1.50	.029322	.28785	.32432
2.00	.025749	.25277	.43243
2.50	.021552	.21157	.54054
3.50	.015994	.15701	.75675
5.00	.011570	.11358	1.0811

Run #3 - 50 rpm

<u>t</u>	<u>C</u>	<u>C/C₀</u>	<u>vt/V</u>
.50	.067831	.66587	.10810
1.00	.058757	.57680	.21622
1.50	.051611	.50665	.32432
2.00	.045485	.44651	.43243
3.00	.035844	.35187	.64865
4.50	.029662	.29118	.97297
6.50	.021552	.21157	1.4054
9.00	.016742	.16435	1.9459

Run #4 - 50 rpm

<u>t</u>	<u>C</u>	<u>C/C₀</u>	<u>vt/V</u>
.50	.058246	.57178	.10810
1.50	.036921	.36244	.32432
2.50	.030626	.30064	.54054
3.50	.026146	.25666	.75675
5.00	.020531	.20155	1.0811
6.50	.016731	.16424	1.4054
8.50	.015154	.14876	1.8378
10.0	.013453	.13206	2.1622

Run #5 - 100 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.047584	.46885	.10810
1.00	.036722	.36182	.21622
1.50	.031817	.31349	.32432
2.50	.022889	.22553	.54054
3.50	.019453	.19167	.75675
4.50	.015721	.15490	.97297
6.00	.014338	.14127	1.29729

Run #6 - 100 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.078267	.77117	.10810
1.00	.069646	.68623	.21622
1.50	.062954	.62029	.32432
2.50	.051667	.50908	.54054
3.50	.047187	.46494	.75675
4.50	.041799	.41185	.97297
6.00	.031760	.31293	1.29729
7.50	.022516	.22185	1.6216
9.00	.016833	.16586	1.9459

Run #7 - 100 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.062613	.61693	.10810
1.00	.045315	.44649	.21622
2.00	.039417	.38838	.43243
3.00	.036297	.35764	.64865
4.00	.034256	.33753	.86486
5.50	.026032	.25650	1.1892
7.00	.019260	.18977	1.5135

Run #8 - 100 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.078890	.77366	.10810
1.00	.077359	.75864	.21622
1.50	.075601	.74140	.32432
3.00	.052007	.51243	.64865
4.50	.044067	.43216	.97297
6.00	.027450	.26919	1.2973
7.50	.019760	.19378	1.6216
9.00	.014201	.13927	1.9459

Run #9 - 200 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.093977	.92161	.10810
1.00	.083881	.82260	.21622
2.00	.067037	.65742	.43243
3.50	.048094	.47165	.75675
5.00	.034086	.33427	1.0811
7.00	.021960	.21535	1.5135

Run #10 - 200 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.094260	.92439	.10811
1.50	.076225	.74752	.32432
2.50	.060401	.59234	.54054
3.50	.048378	.47439	.75675
4.50	.038056	.37321	.97297
6.00	.027563	.27030	1.2973
8.00	.017763	.17419	1.7297

Run #11 - 700 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.092899	.91104	.10810
1.50	.074580	.73139	.32432
2.50	.059948	.58790	.54054
3.50	.047867	.46942	.75675
5.00	.034596	.33927	1.0811
7.50	.020009	.19622	1.6216

Run #12 - 700 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.093183	.91383	.10810
1.50	.075601	.7414	.32432
2.55	.059834	.58678	.55136
5.50	.031590	.30979	1.1892
7.00	.022913	.2247	1.5135
8.50	.016357	.16041	1.8378

Run #13 - 1250 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.087472	.90247	.10810
1.50	.070918	.73168	.32432
2.50	.057275	.59092	.54054
4.00	.041232	.42540	.86486
6.00	.026364	.27200	1.2973
8.00	.017525	.18081	1.7297
10.00	.011138	.11491	2.1622

Run #14 - 1250 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
1.00	.078172	.80652	.21621
2.00	.063713	.65735	.43243
3.50	.045371	.46810	.75615
5.50	.029123	.30047	1.1892
7.50	.018904	.19504	1.6216
9.50	.012139	.12524	2.0540

Run #15 - 2000 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.087523	.90300	.10810
1.50	.070662	.72904	.32432
2.50	.055947	.57722	.54054
4.00	.041079	.42382	.86486
6.00	.026875	.27728	1.2973
8.00	.016963	.17501	1.7297
9.50	.012364	.12756	2.0540

Run #16 - 2000 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.086909	.89666	.10810
1.00	.078632	.81127	.21622
2.00	.062896	.64892	.43243
3.50	.045217	.46652	.75675
5.50	.029583	.30522	1.1892
7.50	.018700	.19293	1.6216
9.50	.012160	.12546	2.0540

Run #17 - 3000 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.087829	.90616	.10810
1.50	.070151	.72377	.32432
2.50	.056100	.57880	.54054
4.00	.040517	.41802	.86486
6.00	.025955	.26778	1.2973
8.00	.016605	.17132	1.7297
9.50	.012507	.12904	2.0540

Run #18 - 3000 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
1.00	.078786	.81286	.21622
2.00	.062538	.64522	.43243
3.00	.050122	.51712	.64865
4.50	.036838	.38007	.97297
6.50	.023554	.24301	1.4054
8.50	.015184	.15666	1.8378

Run #19 - 150 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.074034	.79859	.10810
1.00	.061721	.63679	.21622
1.50	.062385	.64364	.32432
2.00	.052626	.54296	.43243
3.00	.051450	.53082	.64865
4.50	.041539	.42857	.97297
6.00	.030707	.31681	1.2973
8.00	.018547	.19135	1.7297

Run #20 - 400 rpm

<u>t</u>	<u>C</u>	<u>C/C_o</u>	<u>vt/V</u>
.50	.089260	.92092	.10810
1.50	.070968	.73219	.32432
3.00	.051093	.52714	.64865
5.00	.032342	.33368	1.0811
7.50	.019415	.20031	1.6216

APPENDIX IIIDETERMINATION OF MODEL
FOR LOW RPM RANGE

The determination of a model that fits the F-curve for the low rpm range consisted of the following steps:

The presence of a delay action was recognized in the initial portion of the curve. A plug flow reactor of small volume was introduced to account for the delay.

The large and very sudden drop in outlet solution concentration was explained by a plug flow reactor of very small volume (almost short circuit) through which a large portion of the flow passed.

The stirred tank reactor's objective is to describe the exponential-like fall in outlet concentration for the remainder of the curve.

Dead space was added to complete model reactor volumes to the physical size.

Flow and volume parameters were established from relative size of characteristics listed above.

Finally, the F-curve of the model was compared to that experimentally obtained.

APPENDIX IV

REACTION RUNS DATA

T = 25°C

C = concentration

V = 925 ml

A = sodium hydroxide

B = ethyl acetate

 $\Delta C = C_A - C_B$ Run #1 - 50 rpmRun #2 - 50 rpm $C_{A_0} = .098132$ $C_{B_0} = .105159$ $C_{A_0} = .098132$ $C_{B_0} = .10407$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
4.0	.021472	
8.0	.021680	
11.0	.022043	
16.0	.022199	
19.5	.023080	
23.0	.022147	
26.5	.022562	

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
1.0	.031638	
6.5	.021317	
11.0		+0.002956
13.0	.021784	
16.5		+0.003500
18.0	.021680	
21.0	.021680	
23.5	.022769	

Run #3 - No stirring

$$C_{A_0} = .098132 \quad C_{B_0} = .099173$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
6.0	.020746	
9.0	.019865	
12.5	.022043	
16.5	.020798	
19.5		+.007312
21.0	.020643	
23.5	.022095	
26.0		+.003318
28.0	.021265	

Run #4 - 200 rpm

$$C_{A_0} = .098132 \quad C_{B_0} = .099350$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
2.0	.027230	
4.5	.023132	
7.0	.021525	
11.0	.022562	
14.0		.006050
15.5	.022510	

Run #5 - 200 rpm

$$C_{A_0} = .098132 \quad C_{B_0} = .099000$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
1.7	.028734	
6.5	.022821	
9.5	.023184	
12.5	.022718	
18.0		+.000596

Run #6 - 400 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10465$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
.5	.037262	
3.5	.026542	
7.0	.025011	
9.0		-.003729
11.5	.024387	
14.5	.024898	
20.5	.024784	
23.5	.024557	

Run #7 - 400 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10449$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
.7	.036127	
2.5	.027847	
5.5	.024614	
8.0		-.002539
9.5	.024444	
12.0	.024387	
14.5	.024274	

Run #8 - 700 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10294$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
.5	.037602	
4.5	.025975	
8.0	.024331	
12.0	.024444	
14.0		-.002341
16.0	.024738	
18.5	.024387	
22.0	.024727	

Run #9 - 700 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10285$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
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1.0	.034539	
7.0	.025011	
9.5		-.002142
11.5	.023990	
17.0	.024100	
19.5	.023933	

Run #10 - 3000 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10276$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
----------	----------------------	-----------

.5	.037602	
6.0	.025408	
11.5	.024897	
15.0	.023820	
17.5	.024727	
19.0		-.001887
21.0	.024387	

Run #11 - 3000 rpm

$$C_{A_0} = .10163 \quad C_{B_0} = .10225$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
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1.0	.034086	
9.0	.024387	
11.0	.023820	
13.5		-.00911
15.0	.024274	

Run #12 - 1250 rpm

$$C_{A_0} = .101945 \quad C_{B_0} = .10107$$

<u>t</u>	<u>C_A</u>	<u>ΔC</u>
----------	----------------------	-----------

.5	.038396	
5.5	.026088	
8.5	.025522	
10.5	.024955	
13.5	.025238	
15.5		-.001039
17.0	.025238	

Run #13 - 1250 rpm

$$C_{A_0} = .101945 \quad C_{B_0} = .099861$$

t	C_A	ΔC
.52	.037205	
3.0	.027337	
5.5	.024954	
9.0	.024954	
11.5	.024841	
13.5		-.000414
15.0	.024954	

Run #14 - 3000 rpm

$$C_{A_0} = .101945 \quad C_{B_0} = .099751$$

t	C_A	ΔC
1.0	.034539	
3.0	.027677	
6.0	.025181	
8.5	.024954	
11.0		+0.000478
12.5	.025181	
16.0		-.001487

Run #15 - 2000 rpm

$$C_{A_0} = .101945 \quad C_{B_0} = .101696$$

t	C_A	ΔC
.5	.037545	
2.5	.027904	
5.5	.025522	
8.0	.024728	
10.5		-.000951
12.5	.025068	
14.0	.024500	
20.5	.024841	

Run #16 - 2000 rpm

$$C_{A_0} = .101945 \quad C_{B_0} = .101520$$

t	C_A	ΔC
.5	.038339	
3.0	.028017	
8.5	.024954	
11.0	.024954	
13.0		+0.000478

APPENDIX V

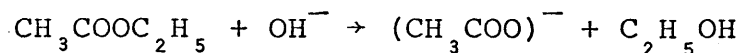
STEADY STATE VALUES FOR COMPLETELY SEGREGATED MODEL

Values tabulated below were obtained from Novosad and Thyn's graphical correlation for conversion limits in a reactor. (38)

Run	C_{A_0}	$R_{(M)} = kT C_{A_0}$	x_M	N_e	R_M/R_S	R_S	$(1-x)_s$	C_A -segregated
1.	.049066	1.640	.488	1.65	1.10	1.492	.465	.0228(5)
2.	.049066	1.640	.485	1.60	1.12	1.465	.470	.0230
3.	.049066	1.640	.471	1.30	1.18	1.390	.500	.0246
4.	.049066	1.640	.470	1.30	1.18	1.390	.500	.0246
5.	.049066	1.640	.469	1.30	1.18	1.390	.500	.0246
6.	.050815	1.698	.482	1.55	1.16	1.465	.485	.0247
7.	.050815	1.698	.481	1.53	1.15	1.477	.480	.0247
8.	.050815	1.698	.476	1.25	1.17	1.450	.492	.0250
9.	.050815	1.698	.476	1.30	1.14	1.490	.492	.0250
10.	.050815	1.698	.476	1.30	1.14	1.490	.492	.0250
11.	.050815	1.698	.474	1.27	1.14	1.485	.498	.0253
12.	.050972	1.703	.470	1.25	1.18	1.442	.500	.0254
13.	.050972	1.703	.466	1.20	1.20	1.420	.505	.0256(5)
14.	.050972	1.703	.466	1.20	1.20	1.420	.505	.0256(5)
15.	.050972	1.703	.466	1.20	1.20	1.420	.505	.0252(5)
16.	.050972	1.703	.471	1.26	1.17	1.456	.498	.0253

APPENDIX VIBASIS FOR CALCULATIONS OF REACTOR CONCENTRATIONS

The chemical reaction used for this study was the saponification or base-catalyzed hydrolysis of ethyl acetate with sodium hydroxide.



The kinetics for this second order, liquid phase, irreversible reaction were obtained from Saldick and Hammett⁽⁴⁴⁾ and from the International Critical Tables⁽⁴³⁾, v. III, p. 129.

The equations derived to give concentration of reactants at steady state or during the approach to steady state were based on material balances. The assumption of equal decrease in reactants concentration was made, which entailed equal-ratio exiting of reactants and of products from the reactor.

The Runge-Kutta method was used to give the numerical solution of the two simultaneous, non-linear, first order differential equations that appear in the approach to steady state of a stirred tank reactor.

A CDC 8090 computer was used to solve the static and transient equations for all models.

APPENDIX VIINOMENCLATURE

C_0	initial concentration
N	number of tanks in series
R	dimensionless reaction number ($k\tau C_0^{q-1}$)
S	stirred tank reactor
T	tubular reactor
q	order of reaction
x	fractional conversion
Δ	concentration difference
τ	reactor residence time
v	volumetric flow rate

Subscripts

M	maximum mixedness
S	complete segregation
e	equivalent (number of tanks)

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