Dynamic Similarity for Imperfect Mixing Processes: Non-Newtonian Fluids

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As production processes for biological and pharmaceutical materials are converted from batch to continuous operation, it becomes necessary to predict the dynamic behavior of the continuous flow systems. Most such processes are carried out in stirred tanks (Malek et al., 1962), and the fluids processed often have non-Newtonian rheological behavior (Deindoerfer, 1960).

Some experimenters report the dynamic behavior of continuous flow mixing systems in which Newtonian fluids are being processed, for example, Cholette and Cloutier (1959, 1968), Manning et al. (1965), Keairns and Manning (1969), and Hubbard and Patel (1971). Analyzing the dynamic response data is carried out in terms of residence time distributions or in terms of mathematical models. Such models have been suggested by the authors cited. The model parameters depend on geometric and process variables. The exact functional dependence must be known to predict the dynamic behavior of a particular system.

Few data are available showing the effects of rheological variables or illustrating scale-up effests in mixing systems in which non-Newtonian fluids are being processed. Bartholomew (1960) reviews some scale-up procedures, and Taguchi et al. (1964) report some such response data. The work of Taguchi illustrates the type of model which has been used to describe the response of continuous flow mixing systems. Results are reported for mixing in a 100 liter cylindrical tank 500 mm in diam. agitated by double impellers 180 mm in diam. A negative step change in feed stream concentration was produced by terminating dye injection at the start of an experiment. Experiments were done using water and using aqueous solutions of carboxymethylcellulose. The rheological characteristics are given in terms of an apparent viscosity which ranged from 50 to 185 cp. The response curves could be approximated by

$$C_o/C_i = 1 \quad \text{for} \quad 0 < Dt < \phi$$

$$C_o/C_i = \exp\left(-\frac{Dt-\phi}{1-\phi}\right) \quad \text{for} \quad Dt > \phi$$

Correlations are presented showing the effects of stirring speed on ϕ for the system used.

$$\phi = K(D/N)^{2/3} \quad \text{for water}$$

$$\phi = K(N)^{-2/3} \quad \text{for CMC solutions} \quad (\mu_{\text{nominal}} < 100 \text{ cp})$$

$$\phi = K(N)^{-\beta} \quad \text{for CMC solutions} \quad (\mu_{\text{nominal}} > 100 \text{ cp})$$

Many other workers using this type of system but differing in inlet and outlet placement and other geometrical details have determined the transient behavior of the output stream when a step change in an input variable is introduced. Often, the variable measured is the concentration of a solute species. If the situation called perfect mixing exists $\overline{C} = C_o$ and the behavior is described by

$$C_o/C_i = \exp(-Dt) \tag{1}$$

This type of system response is observed when the impeller speed is high or when the throughput rate is low. If $\overline{C} \neq C_o$, Equation (1) does not represent the response. The deviation of the system response from the perfect mixing response depends on geometry, on impeller speed, and on feed rate.

Correlating equations for model parameters or other variables depending on the transport phenomena occurring in the system can be developed using a combination of dimensional analysis methods described by Stewart (1965) and Langhaar (1951). This procedure is discussed by Hubbard and Patel (1969). For fluids having rheological behavior described by the power law model, the result of this dimensional analysis is

$$C_o/C_i = f\left(Dt, D/N, \frac{D_i^2 N^{2-\gamma}\rho}{k}, \frac{k}{\rho\,\mathfrak{D}_{im} N^{1-\gamma}}, \frac{N^2 D_i}{g}, \frac{D_T}{D_i}, \frac{H_L}{D_i}, \frac{H_i}{D_i}, \frac{W}{D_i}, \gamma\right) \tag{2}$$

Equation (2) is the basis for developing a correlating equation for any model parameter related to the response of the system. Effects of geometrical ratios can be eliminated by studying geometrically similar systems. Gravitational effects can be made negligible by using baffled tanks and by making the initial solute concentration small enough so that no appreciable density change occurs as the solution is diluted. The effect of N_{Sc} can be neglected if mass transport by molecular interactions is much smaller than mass transport by bulk fluid motion or by turbulent eddy motion. A method for determining the conditions for which this latter assumption is tenable is illustrated below. The Newtonian Schmidt number is the ratio of the kinematic viscosity to the diffusion coefficient—constant in an isothermal system. The impeller speed appears in the non-Newtonian Schmidt number. It cannot be constant if the impeller speed is changed.

When the Schmidt number is neglected, Equation (2) takes the form

$$C_o/C_i = f\left(Dt, D/N, \frac{D_i^2 N^{2-\gamma}\rho}{k}, \gamma\right) \tag{3}$$

Model parameters or other variables can also be correlated

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TABLE 1. DIMENSIONS OF THE CONTINUOUS FLOW MIXING SYSTEMS

Tank volume, liters	D_i, *in.*	D_T/D_i	H_L/D_i	H_i/D_i	H_F/D_i	W/D_i
1.6	2.0	2.5	2.5	1.0	0.5	0.13
5.4	3.0	2.5	2.5	1.0	0.5	0.15
10.9	4.0	2.4	2.4	1.0	0.5	0.14

TABLE 2. RHEOLOGICAL PARAMETERS AND N_{Sc} FOR THE POLYMER SOLUTIONS USED

Mass fraction KELZAN XC	k $\frac{\text{Dyne s}^{\gamma}}{\text{cm}^2}$	γ	$\frac{\rho}{\rho_w}$	Impeller speed (range), s^{-1}	$\frac{k}{\rho N^{1-\gamma}} = N_{Sc}\mathcal{D}_{im}$ (range) (cm²/s)
0.0	0.0175	1.0	1.0	0.42 to 1.67	0.0175
0.0004	8.0	0.24	1.0003	0.15 to 1.67	33.8 to 5.4
0.0068	31.0	0.32	1.006	0.42 to 1.67	55.9 to 21.9

using functions of the form illustrated in Equation (3). The parameter ϕ used by Taguchi et al. could be correlated as

$$\phi = \phi\left(D/N, \frac{D_i^2 N^{2-\gamma}\rho}{k}, \gamma\right) \tag{4}$$

for a power law fluid. The dimensionless time has been omitted in Equation (4), since ϕ is a parameter which represents the entire response curve.

EXPERIMENTAL TECHNIQUES

Experiments were performed in three geometrically similar cylindrical, flat-bottomed tanks equipped with four baffles and agitated by six-blade turbine impellers. The dimensions of each tank are shown in Table 1. The response was determined by a tracer method. Dissolved salt was the tracer. A step change in salt concentration in the inlet stream from 0.1 molar to 0.0 molar was introduced. The salt concentration in the outlet stream was monitored. Details about the construction and operation of the equipment are reported by Calvetti (1971).

To produce non-Newtonian behavior, aqueous solutions of a polysaccharide material (KELZAN XC) were used. Rheological data for the solutions were obtained using a cone-plate viscometer. The Ostwald deWaele model (Bird et al., 1960) was used to describe the fluid behavior. γ, a measure of the deviation from Newtonian behavior, was fixed by the structure and molecular weight of the polymer and was nearly the same for all experiments. k could be varied by changing the concentration of dissolved polymer. Rheological properties of the solutions are listed in Table 2.

EXPERIMENTAL RESULTS

The results show the effect of changing the viscosity parameter k and the effect of changing the system volume while maintaining dynamic similarity. Response data are shown in Figure 1 for solutions containing 0, 0.04, and 0.68% KELZAN XC. For the low feed rates used, significant deviation from perfect mixing occurs only for the most concentrated polymer solution. This indicates that the deviation from perfect mixing may be largely a viscosity effect if $N/D = 0.0018$. Calvetti presents data obtained at different impeller speeds which show the same behavior. The rheological character of the solutions apparently influences the response strongly. For Newtonian liquids, the response can be characterized using only dimensionless momentum transport variables. For non-Newtonian liquids, the Schmidt number should be included in characterizing the response. This makes non-Newtonian data more difficult to analyze than Newtonian data. Since N_{Sc} is proportional to $N^{\gamma-1}$ and N_{Re} is proportional to $N^{2-\gamma}$, the Schmidt number cannot remain constant if the impeller speed is changed to study the effect of Reynolds number. The regime concept described by Johnstone and Thring (1957) must be introduced. In the present work, mass transport by bulk fluid motion is taken to much greater than mass transport by molecular interactions, and the effect of Schmidt number has been neglected. Table 2 shows the range of N_{Sc} for the experiments performed. Since the effective binary diffusion coefficient was not measured, the product $N_{Sc}\mathcal{D}_{im}$ is reported. Data given by Hsu (1967) for diffusion coefficients of ferricyanide ions in hydroxyethylcellulose solutions show that the diffusion coefficient decreases 31% as the polymer concentration is increased from 0% to 1.5%. If the same general behavior

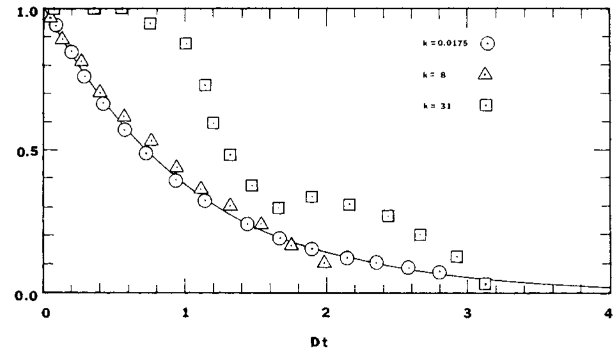

Fig. 1. Typical response data showing the effect of the concentration of dissolved polymer. $N = 25$ rpm, $D_i = 3.0$ in., $D_T = 7.5$ in., $H_i = 3.0$ in., $H_L = 7.5$ in., $F = 240$ ml/min.

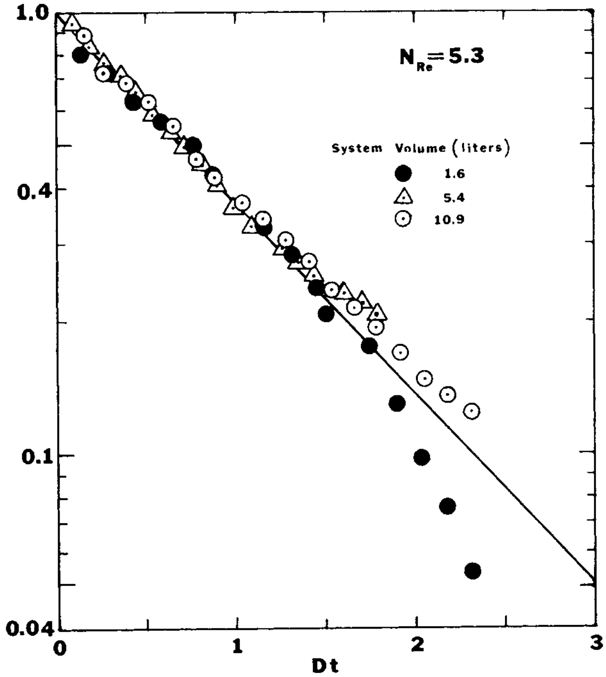

Fig. 2. Effect of system volume. $N_{Re} = 5.3$, $N_{Sc}\mathcal{D}_{im} = 6.4$ to 9.2, $D/N = 0.0018$, $\gamma = 0.24$.

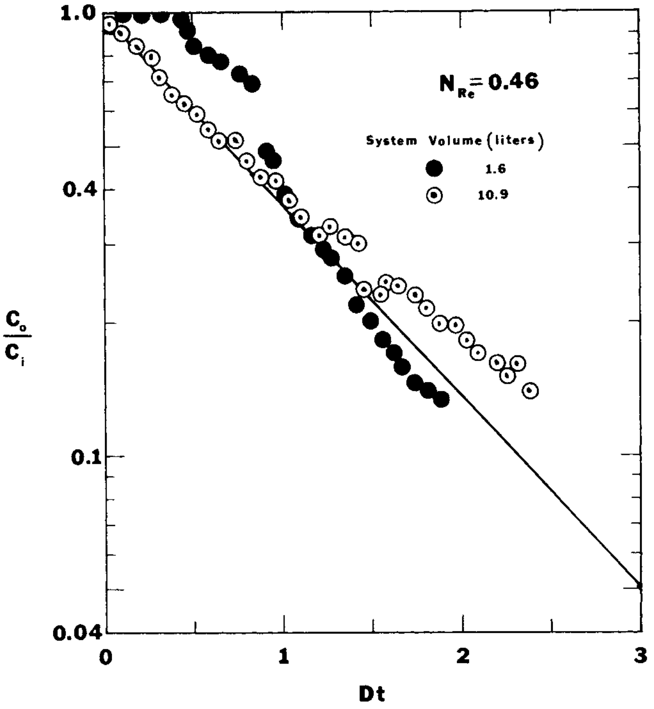

Fig. 3. Effect of system volume. $N_{Re} = 0.46$, $N_{Sc}\mathcal{D}_{im} = 18.4$ to 33.7, $D/N = 0.0018$, $\gamma = 0.24$.

is assumed for the diffusion of salt in the KELZAN XC solutions, the major change in N_{Sc} will come from changes in impeller speed.

Experiments performed in systems having different volumes provide a general method for identifying the regime in which a system is operating. The effect of changing the system volume is shown in Figures 2 and 3. Response data for systems having volumes of 1.6, 5.4, and 10.8 liters are shown for two values of N_{Re}—5.3 and 0.46. Dynamic similarity was maintained for each set of experiments, but diffusive similarity could not be maintained. If the system is operating in a regime in which convection controls the mass transport, then the response curves should coincide when dynamic similarity is maintained no matter what the system volume is. If the curves do not coincide, molecular interactions affect mass transport in some of the systems. Convection alone is not the controlling factor. Figure 2 shows that for $N_{Re} = 5.3$, the response curves coincide. The deviation for $Dt > 1.7$ probably occurs because very low salt concentrations were being measured toward the end of each experiment. Figure 3 shows that for $N_{Re} = 0.46$, the response curves do not coincide. D/N was maintained at 0.0018 for all experiments. It appears that at $D/N = 0.0018$, the system operates in the convective regime only for $N_{Re} > 1.6$.

Correlations formulated according to Equation (4) also show that the system was not always operating in the same regime even though dynamic similarity was maintained. Data for $\phi(N_{Re})$ obtained for $D/N = 0.0018$ falls into two distinct groups—one group for the 0.68% polymer solution and one group for the 0.04% polymer solution.

CONCLUSIONS

Data showing the response to a step change in feed solute concentration obtained for continuous flow mixing systems of different volumes can be used to identify the regime in which the system is operating. This procedure should be useful for any process in which mass transport and momentum transport occur simultaneously.

Maintaining dynamic similarity can be a useful scale-up criterion if both the model system and the prototype system operate in the regime where bulk fluid motion controls the mass transport. Parameters which characterize the system response or the residence time distribution can be correlated in dimensionless form only if all data are obtained for systems operating in the same regime.

NOTATION

$\overline{C}$ = space-averaged solute concentration
C_o = solute concentration at the outlet
C_i = initial solute concentration
D = F/V = dilution ratio
D_i = impeller diameter
D_T = tank diameter
$\mathcal{D}_{im}$ = effective binary diffusion coefficient for solute
F = volumetric feed rate
g = magnitude of gravitational acceleration
H_i = height of impeller above the tank bottom
H_F = height of feed point above the liquid surface
H_L = height of liquid surface above the tank bottom
k = rheological parameter
N = impeller speed
$N_{Re} = \dfrac{D_i^2 N^{2-\gamma} \rho}{k}$ = Reynolds number
$N_{Sc} = \dfrac{k}{\rho \mathcal{D}_{im} N^{1-\gamma}}$ = Schmidt number
t = time
V = volume of tank
W = baffle width
β = exponent (not a constant)
γ = rheological parameter
ρ = fluid density
ϕ = mixing parameter

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