Mixing in Fed-Batch and Continuous Flow Processes in Nonstandard Geometries

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The concept of a standard impeller/tank configuration for mixing, first mentioned in the technical literature by Olney and Carlson (1947), was developed to bring some order to mixing research and to provide geometries in which effective mixing occurred. However, mixing in industrial chemical processing operations is often performed in other configurations. One is to have a centrally-placed flat-blade impeller on the tank bottom, as shown in Figure 1, which is the configuration of interest in this study. Although this geometry may not appear to be the most optimum, very practical process needs require this configuration.

None of the geometries in the mixing studies in the technical literature match the geometry and flow conditions used in this study. The tank used here is part of a fed-batch system, in which the actual chemical processing occurs while the tank is being filled. Mixing is accomplished by the liquid feed jet as well as by the mechanical agitator.

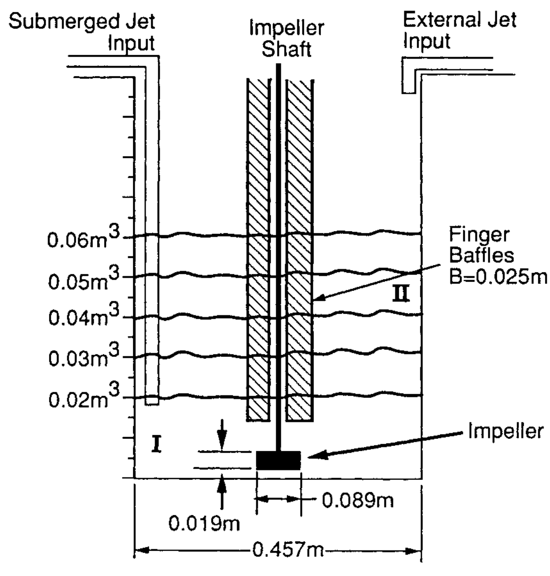

Figure 1. Fed-batch tank geometry.

Table 1. Mixing Time Correlations for Mechanical Agitation

Baffled Tanks Correlations	
1. $N\theta_m = 0.905\ (T/D)^{2.57} \log(2.0/0.05)$	Prochazka and Landau (1961)
2. $N\theta_m = 7.2\ (n_b W/D)^{-0.35}(T/D)^{1.40}(H/D)^{0.5}$	Hiraoka and Ito (1977)
3. $N\theta_m = 5.01\ (T/D)^{2.4}$	Shiue and Wong (1984)
4. $N\theta_m = 2.1\ n_b^{-0.47}(T/W)^{0.74}(T/D)^{1.67}$	Sano and Usui (1985)
Unbaffled Tank Correlation	
5. $N\theta_m = 4.8(T/D)^{0.82}Re^{1/6}$	Hiraoka and Ito (1977)
Jet Based Correlation	
6. $\theta_m = 6\dfrac{H^{1/2}T^{3/2}}{(ND^2)^{2/3}}\dfrac{1}{g^{1/6}}\dfrac{1}{D^{1/2}}$	Norwood and Metzner (1960)

Mechanical Agitation. The mixing time correlations in Table 1 were selected from mixing studies on mechanically agitated batch systems because they have a fairly simple rationale, following Eq. 1:

$$N\theta_m = K(T/D)^a \tag{1}$$

The number of revolutions, $N\theta_m$, to mix a volume depends on some measure of the volume to be mixed and some measure of the impeller size. Impeller geometry is included in some correlations since the ability of an impeller to mix depends on its geometry. The ranges of geometric variables over which the correlations apply have been ignored since they do not match the geometric conditions of this work. An examination of the correlations shows considerable agreement among different exponents.

Jet Agitation. The jet mixing correlations in Table 2 were selected from the literature for their simple rationale. The correlations are for free expanding turbulent jets where the jet Reynolds number is above 2,100. Mixing times in jet-mixed

Table 2. Mixing Time Correlations for Radial Jets in Unbaffled Tanks

1. $\theta_j = 75.4 \dfrac{H^{1/2}T}{(Re)^{1/6}} \dfrac{1}{(U_oD_j)^{2/3}} \dfrac{1}{g^{1/6}}$	Fox and Gex (1956)
2. $\theta_j = 8.7\ T^2/(D_jU_o)$	van de Vusse (1955 a,b)
3. $\theta_j = 5.5\ (D_j/U_o)(T/D_j)^{1.5}(H/D_j)^{0.5}$	Okita and Oyama cited by Maruyama et al. (1982)
4. $\theta_j = 72.4 \dfrac{H^{1/2}T^{3/4}}{(Re)^{0.15}} \dfrac{1}{(U_oD_j)^{0.5}} \dfrac{1}{g^{1/4}}$	Lane and Rice (1981)
5. $\theta_j = 8T^2(Q_oU_o)^{-0.5}$	Fossett and Prosser (1949, 1951)

systems are often calculated from the simple relationship of tank volume divided by an average jet circulation rate, q_{av}, or:

$$\theta_j = V/q_{av} \tag{2}$$

which is the basis for correlations 2, 3 and 5 listed in Table 2. The jet circulation rate as a function of distance in the tank can also be calculated to obtain the average circulation rate (Donald and Singer, 1959; Folsom and Ferguson, 1949; Rushton and Oldshue, 1953,1954,1959,1980). Other correlations for jet mixing times are based on volume and momentum flux of the jet, e.g., correlations 1 and 4 in Table 2. These correlations are similar to the Norwood and Metzner correlation in Table 1.

It should be noted that there is great similarity between Eqs. 1 and 2, and between all the correlations listed in Tables 1 and 2.

Continuous Flow and Fed-Batch Studies. van de Vusse (1955b) discussed continuous mixing in an unbaffled stirred tank. The mixing time, θ_{cf}, for the arrangement was correlated in terms of the mixing time for batch mechanical mixing, θ_m, and the unagitated mixing time, θ_j, due to the feed arrangement and convection occurring in the tank not caused by mechanical agitation. The equation for the mixing time, θ_{cf}, for continuous systems was given as:

$$1/\theta_{cf} = 1/\theta_m + 1/\theta_j + 0.5(\theta_m\theta_j)^{-0.5} \tag{3}$$

This equation matches two expected mixing limits: mixing controlled by jet mixing and mixing controlled by mechanical agitation. A simpler equation might be:

$$1/\theta_{cf} = 1/\theta_m + 1/\theta_j \tag{4}$$

van de Vusse provided data from MacDonald and Piret (1951) and data of his own to justify Eq. 3.

MacDonald and Piret (1951) studied mixing in a continuous flow reactor using dye tracer studies and the continuous hydrolysis of acetic anhydride. In their reactor studies at low mixing levels, the exit concentrations fluctuated unpredictably by as much as 20% above and below the value predicted by the ideal mixing reaction model. In some cases, the degree of conversion for the reaction at the outlet varied to zero as unreacted feed passed through the reactor entirely unmixed.

When considering mechanical and jet mixing in a continuous or a fed-batch process, four possibilities arise:

1. Dominant mechanical and jet mixing
2. Dominant mechanical mixing
3. Dominant jet mixing
4. Neither jet nor mechanical mixing is effective.

When neither jet agitation nor mechanical agitation is adequate, the material may not have a sufficiently high residence time/mixing time ratio, θ_R/θ_{cf}. The ratio has to be on the order of 50 to 200 for effective mixing. Ineffective mixing can also imply that the volume is too large or the distribution of mixing energy is inadequate. Smith et al. (1982) provided an example where liquid jets were inadequate for mixing because the volume was too large. Other examples are readily available in mechanical mixing as well, for example, the impeller is undersized for the volume. For any fixed jet and mechanical mixing conditions, there is a critical volume, beyond which mixing is minimal. The local energy dissipation rate drops to zero outside the critical volume and dead zones are formed. The determination of a critical mixing volume, however, has not been studied extensively.

Objectives

The objectives of this study were to: 1. Obtain mixing times for fed-batch and continuous flow systems; 2. Test the usefulness of the Eqs. 3 and 4 and the correlations given in Tables 1 and 2 for fed-batch and continuous flow systems; 3. Identify the onset of "poor" or less efficient mixing; and 4. Show the general agreement between data and mixing theory.

Experimental Program

Mixing time was considered the time required to disperse a drop of dye uniformly throughout the tank as observed visually. Mixing times were obtained as a function of volume for: 1. batch baffled and unbaffled mechanical mixing; 2. fed jet mixing using different jet configurations; 3. combined jet and mechanical mixing; and 4. combined jet and mechanical mixing while emptying. Mechanical mixing was provided by a radial impeller, and jet mixing was provided by the feed stream. The feed jet discharged vertically downward either above or below the surface. When discharged above the surface, considerable air entrainment occurred.

The tank was 0.457 m in diameter. The liquid height was varied between 0.122 to 0.427 m; liquid volume varied between 0.02 and 0.07 m^3. The impeller was a four-flat-blade impeller, D = 0.089 m, W = 0.0190 m, placed on the tank bottom at minimum clearance. The impeller rotational speed was fixed at 4.0 s^{-1} and the impeller Reynolds number, ND^2/ν, was 31,600 for all studies. Four-finger baffles extended to a height of 0.12 m above the tank bottom and were placed directly above the impeller and symmetrically around the impeller shaft. The tank was fully baffled with no solid body flow behavior.

The feed stream flow rate was constant at 2.33 × 10^{-4} m^3/s and passed through a nozzle, 6.35 mm in diameter. The jet Reynolds number, U_oD_j/ν, was 46,800. The jet discharged vertically downward 50 mm from the wall, either above the liquid surface or as a submerged jet 120 mm from the tank bottom. Air entrainment by the jet was significant. The dye was injected above the liquid with very little momentum; injection time was roughly 2.5 s. The flow behavior was dominated by the local flow conditions at the injection point. Dye was injected at a single location for the mechanical mixing

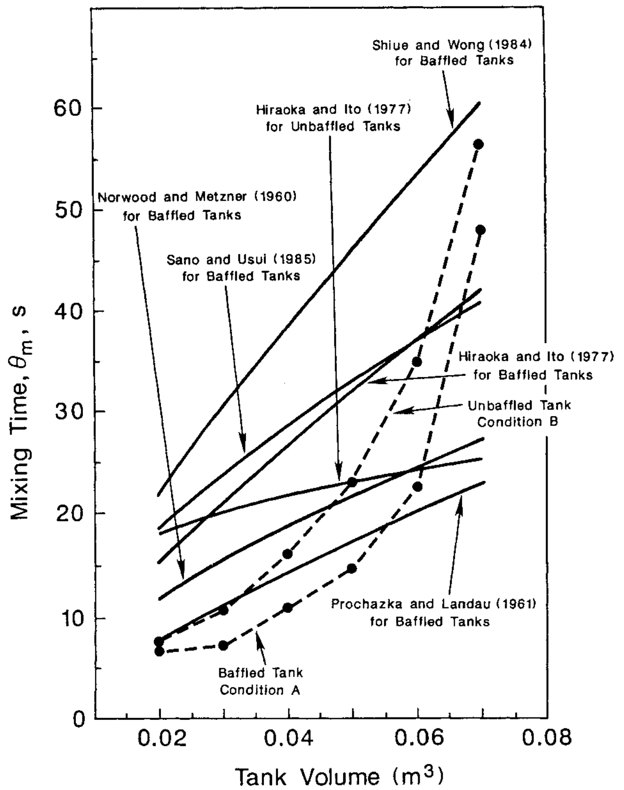

Figure 2. Mixing times obtained from mechanical mixing experiments and predictions from correlations for mechanical mixing.

Conditions: *A*, finger baffles, batch mechanical mixing, no jet mixing; *B*, unbaffled, batch mechanical mixing, no jet mixing

studies but at two positions for jet mixing studies—one away from and the other near the feed point. All locations for the dye injections had the same radial position midway between the impeller shaft and tank wall. Some studies were also performed with the tank being emptied. The output flow was 2.5 $\times$ 10^{-4} m^3/s, which approximately equalled the feed stream flow rate.

The data taken were averages of ten individual mixing times and the mixing times were reproducible. The onset of "poor" mixing occurred in a number of ways. In some cases, dye remained on the surface after injection or became entrapped in a stagnant layer adjacent to a boundary. The standard deviations in mixing times were typically within 10% of the mean, when effective mixing was being accomplished. Standard deviations were higher for the longer mixing times and "poor" mixing conditions.

Results and Discussion

The correlations in Tables 1 and 2 provided rough estimates of the experimentally measured mixing times, as shown in Figures 2 and 3, only when a characteristic length, $(4V/\pi)^{0.333}$, was used as a replacement for both tank diameter and liquid height. When actual liquid height and diameter were used, the correlations were not useful. The observed trends in data in Figures 2 and 3 do not match the trends predicted from the correlations, in which mixing time varies as the power laws of diameter and liquid height. Instead, the mixing time data for

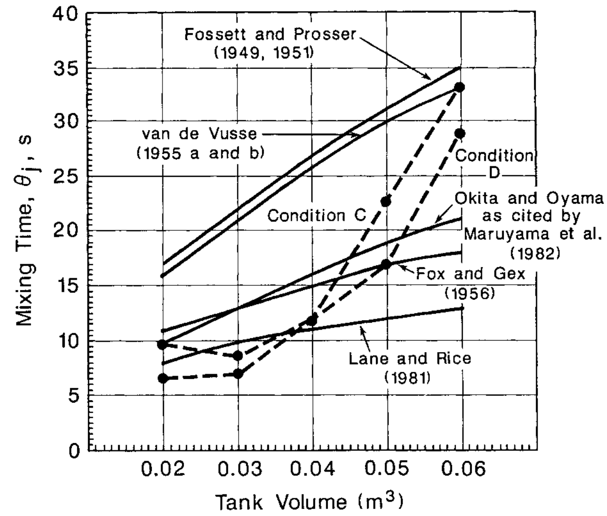

Figure 3. Mixing times obtained from jet mixing experiments and predictions from correlations for jet mixing.

Conditions: *C*, unbaffled, submerged jet mixing, no mechanical mixing, dye injected far away from feed point; *D*, unbaffled, submerged jet mixing, no mechanical mixing, dye injected near feed point

conditions *A*, *B*, *C* and *D* show an exponential relationship with volume or

$$\theta_{cf} = k_1 e^{+k_2 V} \tag{5}$$

The values for k_1 and k_2 as a function of condition are listed in Table 3. k_1 varies with conditions. k_2 is very large and appears to be a constant independent of method of mixing. Figure 4 shows that the mixing data are not inversely additive as expected from Eqs. 3 or 4. However, the data for conditions *E*, *F*, *G* and *H* in Figure 5 show a linear relationship between mixing times and tank volume or

$$\theta_{cf} \alpha V \tag{6}$$

which agrees with Eqs. 1 and 2 and the correlation predictions in Figures 2 and 3.

The data and results appear to be contradictory. Air entrainment and feed location are the only differences between the two groups of data: Group 1, *A*, *B*, *C*, *D*, and *A/D* in Figures 2, 3, and 4; and group 2, *E*, *F*, *G*, and *H* in Figure 5. Group 1 had no air entrainment and group 2 had air entrainment from the surface.

The reason for the differences in mixing behavior can be explained in terms of energy dissipation. Stagnant regions form because bulk circulation and local energy dissipation are in-

Table 3. Constants for the Exponential Relationship in Eq. 5

Condition	k_1 s	k_2 m^{-3}	Regression Coefficient
A	1.72	45.3	0.970
B	3.09	40.9	0.997
C	2.02	46.7	0.983
D	1.80	45.8	0.993

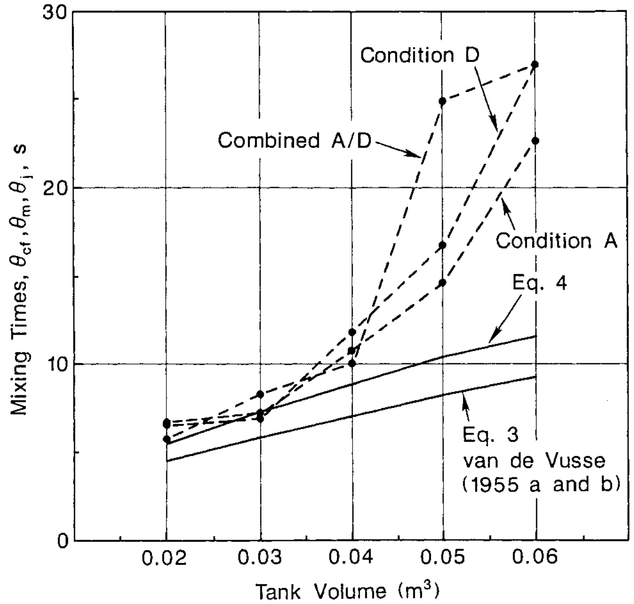

Figure 4. Mixing times obtained from combined mechanical and jet mixing experiments and predictions from Eqs. 3 and 4.

Conditions: *A*, finger baffles, batch mechanical mixing, no jet mixing; *D*, unbaffled, submerged jet mixing, no mechanical mixing, dye injected near feed point; and *A/D*, finger baffles, mechanical mixing, submerged jet mixing, dye injected near feed point

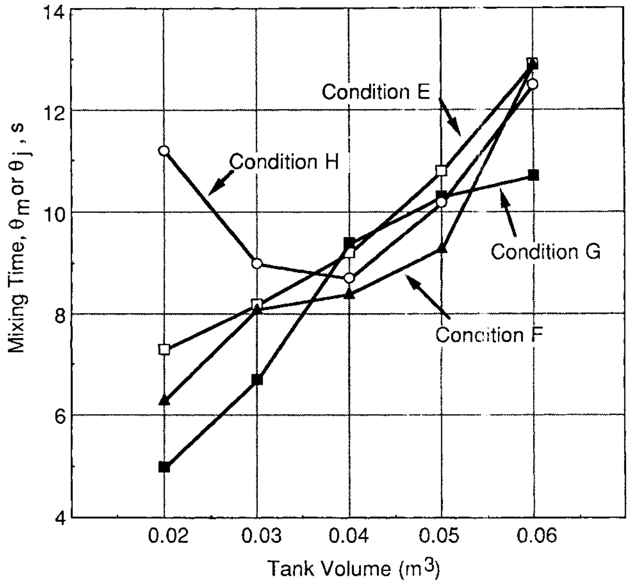

Figure 5. Mixing times for conditions *E*, *F*, *G* and *H*.

Conditions: *E*, finger baffles, mechanical mixing, jet mixing, jet discharge vertically downward above the liquid surface, air entrained in jet; *F*, unbaffled, mechanical mixing, jet mixing, jet discharge vertically downward above the liquid surface, air entrained in jet; *G*, unbaffled, mechanical mixing, jet mixing, jet discharge vertically downward above the liquid surface, tank was being drained, air entrained in jet; *H*, unbaffled, no mechanical mixing, jet mixing, jet discharge vertically downward above the liquid surface, air entrained in jet

sufficient to maintain the local turbulence that accomplishes local mixing. In conditions *A*, *B*, *C* and *D*, the impeller and feed stream were located in one region of the tank labeled as I in Figure 1. Under such conditions, there was insufficient bulk circulation for mixing in the region labeled as II. Without good bulk circulation, Eqs. 1 and 2, and the correlations in the tables are not valid. With the feed jet external to the liquid and located in region II, sufficient circulation was generated by the air entrainment and feed to prevent a stagnant region. The correlations also predict a linear relationship between mixing times and tank volume, as shown in Figures 2 and 3. Overall, the energy input and the distribution of the energy dissipation are important.

The exponential relationship between mixing times and tank volume in Figures 2 and 3 can be explained using work by Rosensweig (1966) and the experimental data of Metzner and Taylor (1960). In a variance balance by Rosensweig, the terms of importance are the time rate of change of the variance and the scalar dissipation function, ϵ_γ. The input and output flows were observed to be unimportant in this work and were dropped from Rosensweig's variance balance. From Rosensweig's work, the variance balance for this work is then:

$$d(\overline{\gamma^2})/dt = -\epsilon_\gamma \tag{7}$$

where ϵ_γ was given as:

$$\epsilon_\gamma = \overline{\gamma^2}\epsilon^{0.33}/\Lambda_\gamma^{0.66} \tag{8}$$

$\overline{\gamma^2}$ is the scalar variance, ϵ is the mechanical energy dissipation rate, and Λ_γ is the scalar integral scale. Substituting Eq. 8 into Eq. 7 and integrating and assuming that $d(\overline{\gamma^2})/(\overline{\gamma^2})$ integrates to a constant which is observed experimentally as the end point of the mixing, an equation for mixing time can be obtained as:

$$\theta = k_3\Lambda_\gamma^{0.66}/\epsilon^{0.33} \tag{9}$$

The last wisp of variance occurred far from either the jet or the impeller and near the outer perimeter of the volume. The conditions there were independent of the mixing method for the most part.

Metzner and Taylor (1960) showed that the energy dissipation data at large distances from the impeller followed an exponential decay with volume from the impeller as:

$$\epsilon = k_4e^{-k_5V} \tag{10}$$

V is a representative volume, and k_4 and k_5 are parameters of the curve fit. The k_5 values approached zero in the data at high impeller rotational speeds. Substituting Eq. 10 into Eq. 9,

$$\theta = k_6\Lambda_\gamma^{0.66}e^{+k_5V} \tag{11}$$

As Eq. 11 indicates, two possible relationships exist for mixing times. When poor mixing occurs, mixing times become an exponential phenomena of volume, and the parameter, k_5, is very large under such conditions. When effective mixing occurs, k_5 becomes very small and the exponential relationship disappears. Typically, the scalar integral scale, Λ_γ, varies linearly with some power of volume.

Equations like Eqs. 3 and 4 fail because they do not apply to nonlinear systems.

Typically, mixing times are independent of injection point as is stated in many mixing studies. Mixing in such systems, however, is usually excellent which essentially causes independence. In "poorly" mixed systems, mixing times will depend on the geometry of the feed locations.

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Notation

a = exponent
D = impeller diameter
D_j = initial jet diameter
g = acceleration of gravity
H = liquid height
k_i = parameters or constants
N = impeller rotational speed, revolutions/second
n_b = blade number
Q_o = initial volumetric flow rate at jet orifice
q_{av} = average circulation rate
Re = impeller Reynolds number, $\pi ND^2/\nu$ or ND^2/ν, jet Reynolds number, $\pi U_o D_j/\nu$ or $U_o D_j/\nu$
T = tank diameter
t = time
U_o = initial axial or average velocity of jet at orifice
V = tank volume
W = blade width
X = axial distance along jet

Greek letters

θ = mixing time
θ_{cf} = mixing time for continuous and fed-batch systems
θ_j = jet mixing time
θ_m = mechanical mixing time
θ_R = residence time
ρ = density
ν = kinematic viscosity
μ = viscosity
ϵ_γ = scalar dissipation function
γ = scalar concentration deviation
$\overline{\gamma^2}$ = scalar variance
ϵ = mechanical energy dissipation rate
Λ_γ = scalar integral scale

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