Effect of Residence Time Distribution of Gas Bubbles on Slurry Reactor Performance

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The effect of mass-transfer resistance from gas bubble to liquid in three-phase slurry reactors may depend on the residence time distribution (RTD) of the bubbles. It has been shown (Niiyama and Smith, 1976) that for very soluble, or slightly soluble, gaseous reactants there is no effect. For very soluble gases, equilibrium is attained shortly after the bubbles leave the dispersion tube. The subsequent path (and RTD) of the bubbles, whether they rise in plug flow through the slurry or remain in the slurry for a long time, is unimportant. For slightly soluble reactant gases, the concentration does not change significantly as the bubbles move through the liquid, and again the RTD is inconsequential. However, for intermediate solubilities the effect has been unknown. This paper presents a quantitative method for determining the influence of the RTD on slurry reactor performance in terms of the pertinent parameters. A criterion of performance is the amount of mass transfer from gas bubble to liquid, as measured by the ratio $C_{g,e}/C_{g,f}$ of reactant concentrations in the effluent and feed gas streams. To characterize the RTD of the bubbles, we use the stirred-tanks-in-series model where for an infinite number of tanks ($n = \infty$) the bubbles are in plug flow, and for $n = 1$ the RTD of complete mixing is obtained. The density $\rho_{s,L}$ of catalyst particles, the concentration C_L of reactant, and the temperature are assumed to be uniform throughout the slurry liquid. Furthermore, the concentration C_L is presumed not to change significantly with time during the average residence time (usually of the order of 1.0 s) of the gas bubbles.

Under these conditions, the mass balance of reactant in the gas phase, for the first tank in the series, is

$$\frac{Q_g}{V_L/n}(C_{g,1} - C_{g,f}) = -k_L a_b \left(\frac{C_{g,1}}{H} - C_L\right) \quad (1)$$

where H = Henry's law constant. Equation 1 may be rearranged to the form

$$C_{g,1} = \frac{C_{g,f} + HC_L(Ab/n)}{1 + (ab/n)} \quad (2)$$

Here Ab is a dimensionless absorption parameter defined as

$$Ab = \left(\frac{k_L a_B}{H}\right)\frac{V_L}{Q_g} = \frac{L/\bar{v}_B}{V_B H/(k_L a_B)} \quad (3)$$

The first equality expresses Ab in terms of easily measurable quantities. The second equality of Eq. 3 shows that Ab may be regarded as a characteristic residence time of the gas bubbles divided by a characteristic time for absorption; $\bar{v}_B$ is the vertical component of the bubble velocity. Equation 2 can be extended to give the concentration leaving the nth tank, by the same procedure as used for homogeneous reactions (Smith, 1981), to yield:

$$\frac{C_{g,e}}{C_{g,f}} = \frac{1}{(1 + Ab/n)^n} + \frac{HC_L}{C_{g,f}}\left[1 - \frac{1}{(1 + Ab/n)^n}\right] \quad (4)$$

Equation 4 gives the effect of the RTD of the gas bubbles on the effluent concentration in terms of n and the concentration C_L of reactant in the liquid phase. This latter concentration is determined by the process of mass transfer form liquid to particle and reaction and diffusion within the catalyst particle. For first-order irreversible reactions an explicit solution can be readily obtained in the following way. Over the whole system of reactors, allowing for steady-state continuous flow of liquid and gas, the mass balance of reactant is

$$\frac{Q_L}{V_L}(C_L - C_{L,f}) + (k_s a_s)(C_L - C_s) = \frac{Q_g}{V_L}(C_{g,f} - C_{g,e}) \quad (5)$$

The concentration C_s at the other surface of the catalyst particles is related to the reaction rate by the expression

$$(k_s a_s)(C_L - C_s) = k\eta\rho_{s,L}C_s \quad (6)$$

where η is the effectiveness (a constant for an isothermal first-order reaction) for the liquid-filled pores of the particles. If Eq. 6 is solved for C_s and the result substituted in Eq. 5 the following expression is obtained for C_L:

$$C_L = \frac{Q_L/V_L}{Q_L/V_L + k'}C_{L,f} + \left(\frac{Q_g}{V_L}\right)\frac{1}{Q_L/V_L + k'}(C_{g,f} - C_{g,e}) \quad (7)$$

where k' is a composite, liquid-particle mass transfer and reaction-rate constant

$$k' = \frac{k\eta\rho_{s,L}}{1 + k\eta\rho_{s,L}/(k_s a_s)} \quad (8)$$

Equation 7 can be simplified for most practical cases. For example, if the feed liquid contains no reactant, $C_{L,f} = 0$ and

$$C_L = \frac{Q_g}{V_L}\left(\frac{1}{Q_L/V_L + k'}\right)(C_{g,f} - C_{g,e}) \quad (9)$$

Also, for a batch process with respect to the liquid phase $Q_L = 0$, and Eq. 7 becomes

$$C_L = \frac{Q_g}{V_L k'}(C_{g,f} - C_{g,e}) \quad (10)$$

In terms of a Damkoehler-type number, defined as

$$Da = (Q_L/V_L + k')/(k_L a_B), \quad (11)$$

both Eqs. 9 and 10 may be written

$$HC_L = \left(\frac{1}{Da}\right)\left(\frac{1}{Ab}\right)(C_{g,f} - C_{g,e}) \quad (12)$$

Finally, substitution of Eq. 12 into Eq. 4 gives

$$\frac{C_{g,e}}{C_{g,f}} = \frac{(1 + Ab/n)^n + (Ab)(Da) - 1}{(1 + Ab/n)^n[1 + (Ab)(Da)] - 1} \quad (13)$$

Equation 13 shows that the effect of RTD (via n) on $C_{g,e}/C_{g,f}$ depends on two parameters, Ab and Da. Figure 1 is a plot of Eq. 13 for $Da = 1.0$. For very soluble gases (very small H) Ab becomes very large and $C_{g,e}/C_{g,f} \rightarrow 0$, regardless of the value of n. Similarly, for slightly soluble gases $Ab \rightarrow 0$ and again the RTD of the gas bubbles has no effect. The greatest effect of RTD is given by the relative difference Δ, between results for plug flow and $n = 1$: that is,

$$\Delta = \frac{(C_{g,e})_{n=1} - (C_{g,e})_{n=\infty}}{C_{g,f} - (C_{g,e})_{n=\infty}} \quad (14)$$

The Δ curve in Figure 1 shows that this difference is significant at $Da = 1.0$ and has its maximum, $\Delta = 0.17$, at an intermediate value of $Ab = 2.2$. As Da increases, which corresponds to relatively more resistance of gas-to-liquid mass transfer, the effect of RTD increases. For example, at $Da = 10$, the maximum value of Δ is 0.22 and this occurs for $Ab = 1.8$. For Da approaching zero there is no

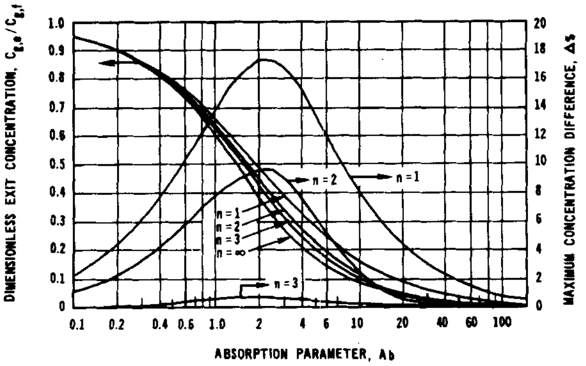

Figure 1. Effect of RTD on Exit Gas Concentration for Da = 1.0

bubble-to-liquid mass transfer resistance and $\Delta = 0$; the effect of RTD of the gas bubbles disappears, regardless of Ab.

Equation 14 is equivalent to a difference in conversion between $n = 1$ and $n = \infty$ divided by the conversion for $n = \infty$. Hence, it should be useful for deciding whether the RTD of the gas bubbles will have an effect on reactor performance or on interpreting laboratory, slurry-reactor data. The procedure is to estimate values of Ab and Da and then calculate Δ from Eq. 14, obtaining the $C_{g,e}/C_{g,f}$ values from Eq. 13. This will give the maximum effect of RTD. It is highly unlikely that the RTD will approach that of one stirred tank. A more realistic measure of the effect of RTD would be a comparison of $C_{g,e}/C_{g,f}$ for $n = \infty$ and $n = 2$ or 3. For these conditions Figure 1 shows that the error would be much less than the maximum values given in the previous paragraph.

ACKNOWLEDGMENT

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NOTATION

Ab	= Absorption parameter, defined by Eq. 3
C_g	= concentration of reactant in gas phase, kmol/m^3; $C_{g,e}$ = concentration in effluent gas; $C_{g,f}$ = concentration in gas feed
C_s	= concentration of reactant in the liquid at the outer surface of the catalyst particle, kmol/m^3
C_L	= concentration of reactant in liquid phase, kmol/m^3; $C_{L,f}$ = concentration in liquid feed
Da	= Damkoehler number, defined by Eq. 11
H	= Henry's law constant for reactant $H = (C_g/C_L)_{\text{equil}}$
k	= first-order reaction rate constant m^3/(s)(kg catalyst)
k'	= composite rate constant defined by Eq. 8, s^{-1}
$(k_L a_B)$	= volumetric mass transfer coefficient, gas-bubble to liquid, (m/s)[m^2/(m^3 of bubble and particle-free liquid)]
$(k_s a_s)$	= volumetric mass transfer coefficient, liquid-to-particle, (m/s)[m^2/(m^3 of bubble- and particle-free liquid)]
L	= height of liquid in slurry reactor, m
n	= number of well-stirred tanks in series
Q_g	= volumetric flow rate of gas through reactor, m^3/s
Q_L	= volumetric flow rate of liquid in or out or reactor, m^3/s
RTD	= residence time distribution of the gas bubbles
V_B	= gas-phase holdup in slurry, m^3 of gas/(m^3 of bubble- and particle-free liquid in reactor)
$\bar{v}_B$	= average vertical component of bubble velocity, m/s
V_L	= volume of bubble- and particle-free liquid in the reactor, m^3

Greek Letters

Δ	= fractional conversion difference, defined by Eq. 14
$\rho_{s,L}$	= density of catalyst particles in the liquid, kg of particles/(m^3 of bubble- and particle-free liquid)
η	= effectiveness factor for reaction in the catalyst particles

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