THE EFFECT OF IMPERFECT MIXING ON GAS-LIQUID REACTIONS: LINKAGE BETWEEN LIQUID-FILM AND BULK LIQUID

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ABSTRACT

The extensive literature on gas-liquid reactions has, with a few exceptions, been devoted to reactions that are considered to be completed entirely in liquid-film or bulk liquid for the sake of simplification of modeling and design. A different perspective for modeling of gas-liquid reactions is presented here. The two-way linkages between liquid-film and bulk liquid, largely ignored previously, are properly accounted for in the present approach. This is especially essential in situations where the gas and liquid phases are not perfectly mixed (situations very frequently encountered in industrial operations), since the composition of the liquid in the film and the bulk both vary with axial position in the reactor. A first-order irreversible reaction, a perfectly mixed liquid phase, and non-perfect mixing in the gas phase are considered as an example. The effects of transport and kinetic parameters on reactor performance are presented via appropriate numerical illustrations. An unwarranted confinement of the liquid phase reaction to either portion of the liquid phase is shown to lead to incorrect design and erroneous prediction of performance of gas-liquid reactors.

Introduction

The interactions between mass transfer and chemical reaction require simultaneous consideration of molecular diffussion, fluid dynamics, and reaction kinetics. Molecular diffusion is a form of micromixing in which the mixing process is viewed in terms of the intimacy of mixing of molecules in a flow system. Another mixing phenomena is macromixing that considers the fluid as independent entities and provide information on the residence time experienced by each. For describing macroflow effects in continuous reactors the concepts of residence time distribution, backmixing and axial dispersion are used.

In this paper, the bulk gas phase is assumed to be non-perfectly mixed, and the bulk liquid phase is assumed to be perfectly mixed. The macromixing model used to model the bulk gas phase is the single parameter axial dispersion model. For the extreme situation where there is no mixing in the gas phase, the plug flow model is assumed. Work on a single reaction and multiple reactions for a perfectly mixed bulk

gas and liquid phases were considered recently (Parulekar and Saidina Amin, 1996a and b, Saidina Amin, 1996).

The axial dispersion coefficient for an arbitrary species J in the gas phase is expressed in the dimensionless form as the Peclet number ($Pe_{JG} = u_G H_J / D_{JG}$). The value of the Peclet number denotes the degree of backmixing. If $Pe_{JG} = 0$, backmixing is complete, and for $Pe_{JG} = \infty$, plug flow prevails. The model can be applied to represent the bubble column, spray tower and packed column reactors.

Problem Formulation

A mathematical model, in dimensionless form, for a perfectly mixed liquid phase and non-perfectly mixed gas phase is presented. A single liquid phase reaction in which at least the reactant (A) or the product (B) is volatile is considered as an example. A schematic representation of a section of a reactor is shown in Fig. 1. Details of this model are available in Saidina Amin, 1996.
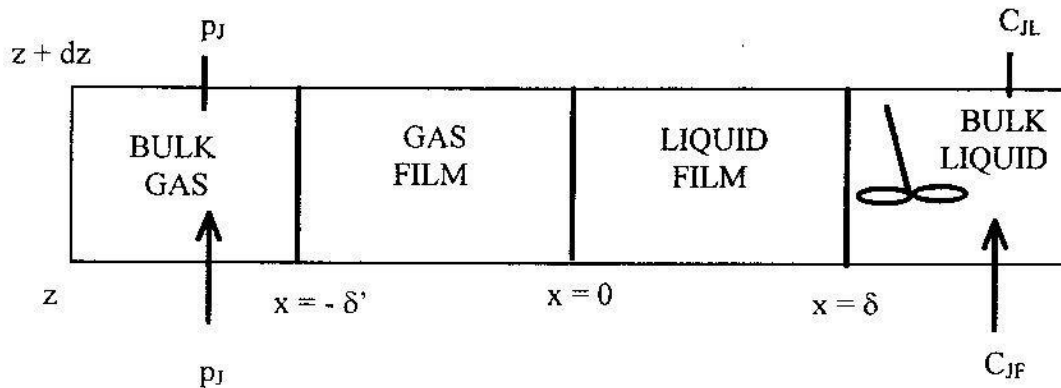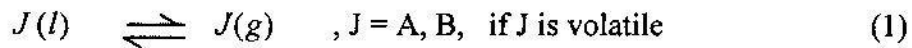
$$J(l) \; \rightleftharpoons \; J(g) \quad , J = A, B, \quad \text{if J is volatile} \tag{1}$$

$$A(l) \; \longrightarrow \; B(l) \tag{2}$$

Fig. 1. A Schematic Representation of The Reactor Model

The dimensionless parameters are stated in eq. 3.

$$J = \begin{cases} \dfrac{C_J}{v_J C_A^*} & \text{liquid phase} \\[4mm] \dfrac{p_J}{v_J H_J C_A^*} & \text{if } H_J > 0 \text{ gas phase} \end{cases} \quad, \quad J_I = \dfrac{C_{JI}}{v_J C_A^*}, \quad J_{LF} = \dfrac{C_{JF}}{v_J C_A^*},$$

$$J_L = \dfrac{C_{Jb}}{v_J C_A^*}, \quad J_{GF} = \dfrac{p_{jF}}{v_J H_J C_A^*} \quad \& \quad J_G = \dfrac{p_J}{v_J H_J C_A^*} \quad \text{if } H_J > 0, \tag{3}$$

$$\alpha_0 = \dfrac{D_{AL}}{D_{BL}}, \quad \Re = \dfrac{\delta^2 r}{D_{AL} C_A^*}, \quad q_L = \dfrac{a \varepsilon_L^\tau L^r b}{C_A^*}, \quad \eta = \dfrac{a \varepsilon_L^\tau L D_{AL}}{\delta}, \quad y = \dfrac{x}{\delta},$$

$$y' = \dfrac{z}{H}, \quad Pe_{JG} = \dfrac{u_G H}{D_{JG}}, \quad m_J = \dfrac{H_J}{RT}, \quad \varphi_J = \dfrac{k_{JG} RT \delta}{D_{JL}}, \quad J = A, B$$

The species conservation equations for the liquid-side film, gas-liquid interface (y = 0), the bulk liquid and bulk gas phases can be stated as

$$\dfrac{d^2 A}{dy^2} = \Re \quad \& \quad \dfrac{d^2 B}{dy^2} = -\alpha_0 \Re, \quad 0 < y < 1; \quad J(0) = J_I, \ J(1) = J_L, \ J = A, B \tag{4}$$

$$-\dfrac{1}{\varphi_J} \dfrac{dJ}{dy} = m_J \left(J_G - J_I \right) \quad \text{at } y = 0 \tag{5}$$

$$A_{LF} - A_L - q_L - \eta \dfrac{\overline{dA}}{dy} \bigg|_{y=1} = 0, \qquad B_{LF} - B_L + q_L - \dfrac{\eta}{\alpha_0} \dfrac{\overline{dB}}{dy} \bigg|_{y=1} = 0 \tag{6}$$

with

$$\dfrac{\overline{dJ}}{dy} = \int\limits_0^1 \dfrac{dJ}{dy} dy', \qquad J = A, B. \tag{7}$$

$$m_A \left(\dfrac{1}{Pe_{AG}} \dfrac{d^2 A_G}{dy'^2} - \dfrac{dA_G}{dy'} \right) + \dfrac{\tau_G}{\tau_L} \eta \dfrac{dA}{dy} \bigg|_{y=0} = 0,$$

$$\tag{8}$$

$$m_B \left(\dfrac{1}{Pe_{BG}} \dfrac{d^2 B_G}{dy'^2} - \dfrac{dB_G}{dy'} \right) + \dfrac{\tau_G}{\tau_L} \dfrac{\eta}{\alpha_O} \dfrac{dB}{dy} \bigg|_{y=0} = 0$$

Numerical Illustration

The parameter values used in the numerical illustrations to be discussed next are based on recommendations in prior literature (Doraiswamy and Sharma, 1984; Westerterp *et al.*, 1984; Landau, 1992; Winkelman and Beenackers, 1993). In all simulations, the kinetic coefficient is considered to be the variable parameter. All numerical illustrations pertain to the situation with A and B being volatile.

The parameters employed were

$n = 1$, $A_{GF} = 1$, $B_{LF} = B_{GF} = 0$, $m_A = 0.1$, $m_B = 0.01$, $\alpha_0 = 2$, $\eta = 20$,
$\alpha_A = 150$, $\alpha_B = 300$, $\tau_G = 0.1\tau_L$, $\alpha = 0.999$.

The relationship between k and γ is

$$\gamma = \sqrt{\frac{k\delta^2}{D_{AL}}}$$

Fig. 2 indicates the concentration profiles of species B in the liquid and gas phases respectively, B_L and B_G, with axial positions as k (or γ) increases. At low k (or γ), B_G increases with axial position and approaches B_L, which is independent of the axial position, towards the end of the column. As k increases, B_L and B_G approach near equilibrium. With further increase in k, there is a reversal in the direction of mass transfer and the occurrence of the reversal becomes more critical with higher k. The species B is observed to be locally absorbed instead of being desorbed in some portions of the reactor. Globally, B is still being desorbed as indicated by the equation from the mass balance

$$m_B B_G < B_L.$$

With increasing k, only a small portion of the reactor is used to strip the species B. The design of the reactor can be improved if the portion that absorps the species B is eliminated. The profiles of B with increasing axial positions for different k for the situation where the reaction is relegated to only the bulk liquid *did not* indicate any reversal in the direction of mass transfer (Fig. 3). These illustrations emphasize the risk of confining the reaction to the bulk liquid only for the sake of simplification in reactor design. Similar patterns are also observed when the mass transfer resistance in the liquid phase is absent.

The conversions of A, f_A, for the three cases under consideration (i. e. reaction in the whole liquid phase, negligible reaction in liquid-film, negligible liquid phase mass transfer resistance) are compared at a particular k that corresponds to γ = 1.0 When $Pe_{AG} = Pe_{BG} = 1$, the values of f_A by considering reaction in liquid-film and by neglecting reaction in liquid-film are 0.984 and 0.978, respectively. When the gas and liquid phases are well-mixed, the corresponding values of f_A by considering reaction in liquid-film and by neglecting reaction in liquid-film are 0.933 and 0.923, respectively. The values of f_A for negligible diffusional resistance in the liquid phase is 0.988 when $Pe_{AG} = Pe_{BG} = 1$ and 0.968 when the gas and liquid phases are well-

mixed. The considerable potential for better utilization of the reactant A by reducing the liquid phase mass transfer resistance is evident from these results. The differences in the conversions of A for the three cases under consideration indicate mixing hurts the performance of a gas-liquid reactor. The portraits of B_L and B_G with the Damkohler number (Da $= \varepsilon_L \tau_L k$) for different degrees of mixing or different Pe_{AG} are shown in Fig. 4. The portraits demonstrated the effect of mixing on the concentrations of species B with increasing k when the reaction is considered to occur in the entire liquid phase and there is diffusional resistance in the liquid phase.

Conclusions

The modeling of gas-liquid reaction considering linkages between the liquid-film and bulk liquid is considered here. The bulk gas phase is non-perfectly mixed while the bulk liquid phase is perfectly mixed. A first-order irreversible reaction was considered for further analysis. The degrees of mixing in the gas phase is shown to hurt the performance of the gas-liquid reactor. The numerical illustrations revealed that the assumption of confining the reaction to only one part of the reaction phase could lead to erroneous design and prediction of performance of gas-liquid reactors.

Acknowledgment

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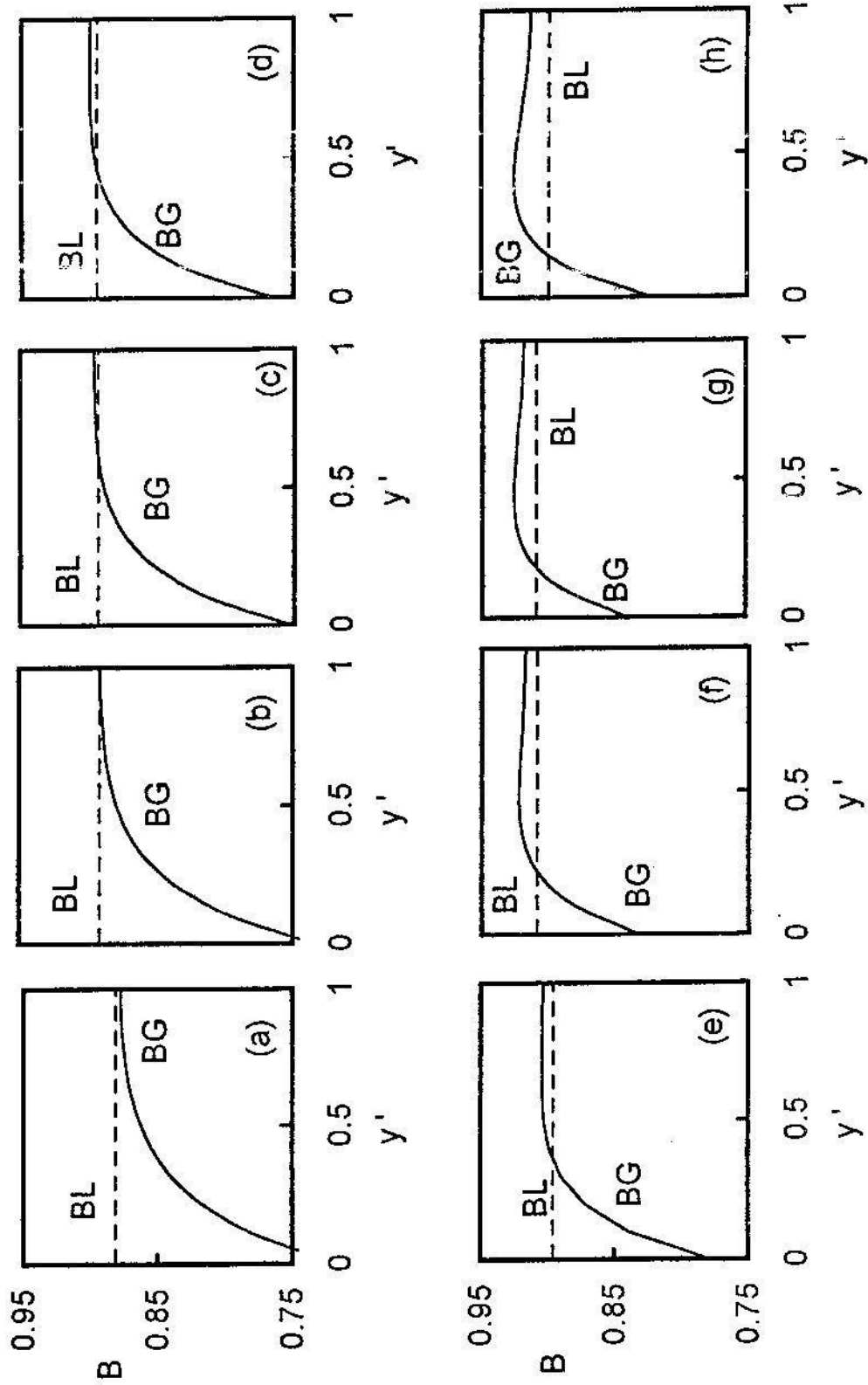
Fig. 2. Profiles of B_L and B_G with Axial Positions for Irreversible Reaction For A and B Volatile. The values of γ are (a) 0.1, (b) 0.5, (c) 0.8, (d) 1.0, (e) 1.2, (f) 1.5, (g) 1.8, (h) 2.0. $Pe_{AG} = Pe_{BG} = 1$.
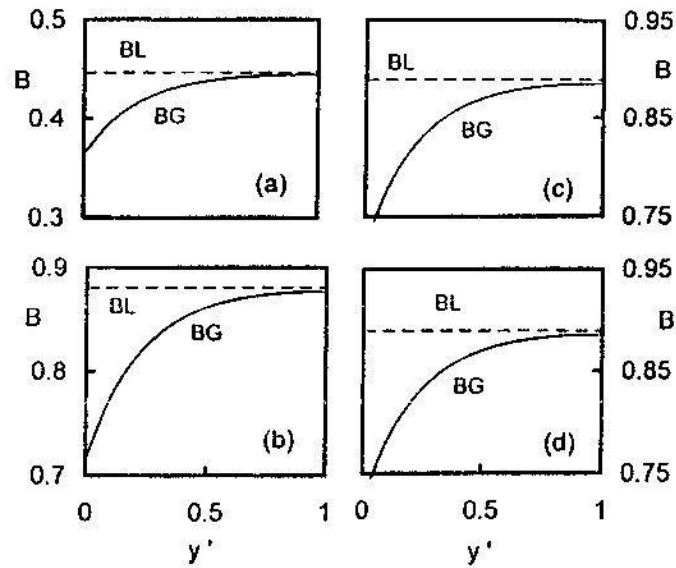
Fig. 3. Profiles of B_L and B_G with Axial Positions for Irreversible Reaction For A and B Volatile When Reaction in Liquid-Film is Negligible. The values of γ are (a) 0.01, (b) 0.1, (c) 1.0, (d) 2.0. $Pe_{AG} = Pe_{BG} = 1$.
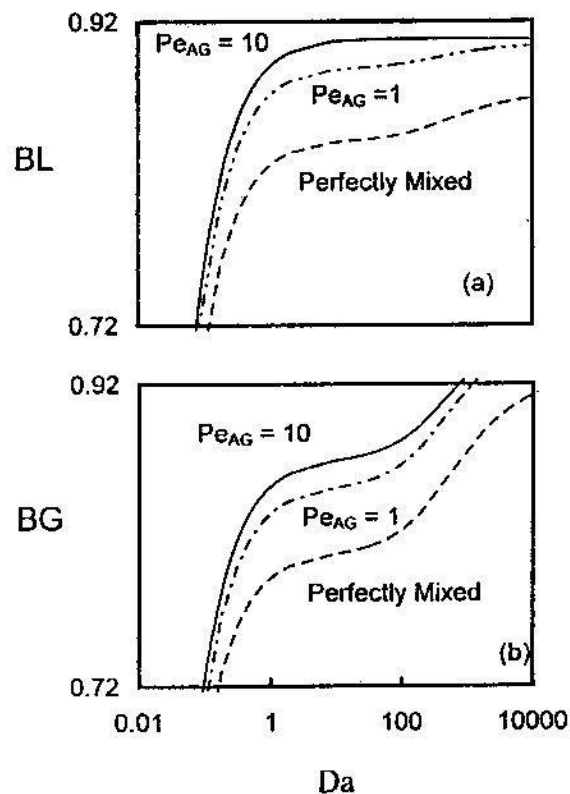

Fig. 4. Profiles of B_L and B_G with Damkohler Number for Different Fluid Flow Patterns