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BEHAVIOUR OF SUCCESSIVE CONTACT MODEL FOR CATALYTIC REACTION IN FLUID BEDS*

TERUKATSU MIYAUCHI

Department of Chemical Engineering, University of Tokyo, Tokyo

In a companion paper (*J. Chem. Eng. Japan*, **7**, 201, 1974), a concept of the successive contact model has been presented to account for the extent of catalytic reaction in a fluid bed. The model has been tested here for its soundness by comparing the behaviour of model with catalytic reaction data available so far.

An apparent fraction of catalyst μ^* in the bubble phase for catalytic hydrogenation of ethylene is shown to be reasonably well estimated from catalyst distribution in the dilute phase. The model provides a sound explanation for an apparent anomaly of μ^* being fairly greater than unity in some data. Influence of bed design and operation on selectivity of a given catalytic reaction is briefly given for fluid beds.

1. Introduction

In a companion paper¹⁴⁾ a concept of successive contact model has been introduced to take into consideration the contribution of dilute phase catalyst to overall conversion of catalytic reaction. The reason of introducing this model was primarily to avoid the difficulty observed by Gilliland and Knudsen⁷⁾ for usual two phase models of fluid beds.

In what follows, the model will be tested for its soundness by comparing its performance with experimental data available so far. Those given by Lewis¹²⁾ and Gilliland⁷⁾ are extensively utilized here, since their reaction data are taken under relatively higher rate of reaction and provide the severest test of models. It is usually hard to distinguish the soundness of one model from another when reaction rate is slow or moderate, since the reaction taking place in the emulsion phase bulk becomes controlling for the progress of overall conversion.

2. Preliminary Consideration

The reaction used by Lewis¹²⁾ was hydrogenation of ethylene catalysed by a nickel-coated cracking catalyst at an average temperature of 113°C under about atmospheric pressure. Reaction occurred with

a large excess of ethylene present, so that the reaction was first-order in hydrogen and the reverse reaction was negligible. The particles fluidized consisted primarily of the 100~140 mesh fraction (average particle diameter $d_p=122\ \mu$) of microspherical Aerocat (MS-A) cracking catalyst. Quiescent bed density was 0.52 g/cc and quiescent gas velocity was 0.73 cm/sec. Some work was also done with two fractions of impregnated alumina of 188 and 80 μ in d_p .

Their fluidized bed consisted of a 4 ft. length of 2.05 in. i.d. Pyrex tubing. Physical parameters they used were mainly those based on the quiescent bed density. According to Morooka *et al.*¹⁸⁾, this bed density is about 1.09 times greater than the emulsion phase density during fluidization for a fluid bed of FCC catalyst (which has the quiescent bed density of 0.54 g/cc). The factor λ ($=1.09$ in the above case) is found¹⁸⁾ to be rather independent of gas velocity and bed diameter. Lewis determined catalyst activity K by a fixed-bed catalyst tester, and correlated their fluid bed data according to the direct contact model of the VUE modification. Based on these, they obtained the fraction of catalyst in the bubble phase, a , and the gas exchange rate, F .

Physical quantities which correspond to K , a , and F are k_r , μ^* and $k_{ob}a_b$, respectively. Relationships between these variables are given as follows:

$$K=\lambda k_r \quad (1)$$

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〒113 東京都文京区本郷7-3-1

東京大学工学部化学工学科 宮内照勝

Fig. 1 Comparison of experimental "a" with calculation based on successive contact model

$$a = \frac{\text{total volume of catalyst in bubble phase}}{\text{total volume of catalyst in whole bed}} \quad (2)$$

$$= \frac{\mu^* \varepsilon_b}{\varepsilon_e + \mu^* \varepsilon_b}$$

$$F = \frac{\text{volumetric rate of gas exchange}}{\text{volume of quiescent emulsion phase}} \quad (3)$$

$$= \lambda \left(\frac{k_{ob} a_b}{\varepsilon_e} \right)$$

To see the applicability of the VUE modification to the data by Lewis, the range of φ and ξ (ref. to Eqs. (15) and (13) of ref. 14) have been estimated in what follows. The partition ratio m of hydrogen was not measured. However, m for FCC catalyst (without nickel impregnation) at room temperature and atmospheric pressure was found as 0.83 when measured by a chromatographic method. Allowing slight adsorption of hydrogen on nickel, m has been taken as unity under the reaction conditions.

Possible approximate numerical values are estimated from the literature for such variables as $k_b^{16,23}$, a_b^{17} , $\varepsilon_e (=1-\varepsilon_b)^{18}$, $k_{ex} a_{ex}^{11,16}$, and k_r^{12} .

The overall coefficient of mass transfer k_{ob} has been calculated from Eq. (A.6) by assuming freely rising bubbles (ref. to Appendix). When the bubbles are retarded by the wall effect, φ values are still higher than those for the freely rising bubbles.

The order of magnitude of variables estimated for operational range of $U_f = 10 \sim 40$ cm/sec is as follows; $k_{ob} a_b \approx 0.35 \text{ sec}^{-1}$, $\varepsilon_e u_{eu}/U_f \approx 1$, $k_{ex} a_{ex} \approx 4 \sim 10 \text{ sec}^{-1}$, $\beta_r \approx 1 \sim 1.5$ and $\varepsilon_e k_r \approx 1 \sim 10 \text{ sec}^{-1}$ for the dense phase. When these values are introduced into Eqs. (15) and (13) of ref. 14, one has $\varphi \approx 25 \sim 38$ and $\xi \approx 0.95 \sim 0.73$ for $U_f = 10$ cm/sec, and $\varphi \approx 52 \sim 63$ and $\xi \approx 0.98 \sim 0.83$ for $U_f = 40$ cm/sec. Thus, the VUE modification is well applicable to Lewis' reaction data, since $\varphi \gg 10$ and ξ shows little influence on k_{or} due to k_{ob} being the controlling factor for it.

3. Test for Data by Lewis, Gilliland and Glass¹²⁾

3.1 Fraction of catalyst, "a"

As defined in ref. 14, μ^* , an apparent volume fraction of catalyst in the bubble phase is related,

through Eq. (2), with the fraction of catalyst in the bubble phase, a , obtained experimentally by Lewis¹²⁾. On the other hand, the actual fraction of catalyst in the bubble phase, μ may usually be negligible in comparison with μ^* for fluid beds without internals, because μ^* calculated by the use of Eq. (2) and the experimentally obtained "a" amounts to 0.44~0.58 for $U_f = 10 \sim 40$ cm/sec and is about two orders greater than experimental μ -values. Under this circumstance, Eq. (17) of ref. 14 is reduced to

$$\mu^* \varepsilon_b = \int_1^{Z_t} \varepsilon_e dZ \quad (4)$$

By introducing Eq. (4) into Eq. (2) and then utilizing a formal relation for the dense phase that

$$\varepsilon_e = \int_0^1 \varepsilon_e dZ, \text{ one has}$$

$$a = \frac{\int_1^{Z_t} \varepsilon_e dZ}{\int_0^{Z_t} \varepsilon_e dZ} = \frac{H_f - H_t}{H_o - H_t} \quad (5)$$

where H_o , H_f , and H_t are the manometer readings at $z=0$, L_f and L_t respectively (ref. to Fig. 2 of ref. 14). The term "a" has been evaluated by utilizing Eq. (5) and Figs. 2 and 3 of ref. 14 for the bed with $d_p = 128 \mu$ and $L_q = 20 \sim 60$ cm. **Figure 1a** compares "a" thus evaluated with that given by Lewis *et al.*¹²⁾ Unexpectedly good agreement is observed between these two. Also, different quiescent bed heights give about the same "a" in accordance with Lewis. This consistency may perhaps be somewhat fortuitous, since there is ambiguity¹⁴⁾ in defining the location of L_f . In spite of this, the successive contact model is seen to provide a reasonable behaviour of the bed in accordance with the reaction performance.

Figure 1b illustrates "a" determined for 85 μ FCC catalyst (Fig. 4 of ref. 14) and that given by Lewis for 80 μ alumina catalyst impregnated with nickel. Agreement is moderate, perhaps due to difference in density between the two catalysts.

In applying Figs. 2~4 of ref. 14 to obtain "a", it has been tacitly assumed that distribution of particles in the dilute phase fluidized by air is equally applicable to the case of ethylene fluidization at the reaction temperature. The terminal velocity of the catalyst particle in air has been estimated as being by about 25% less than that in ethylene, both being in the Stokes regime. Movement of particles in the dilute phase is essentially in mutually hindered motion, so that the influence of gas properties is assumed to tend to diminish.

3.2 Mass transfer term, F

Since the term "a" has shown good agreement between the two models, the term F is necessarily equivalent in the two for 122 μ dia. particles. As shown in **Fig. 2**, $k_{ob} a_b / \varepsilon_b$ has been obtained as a function of U_f , by introducing experimental data¹²⁾ for

F into Eq. (3), where ε_b has been determined* from Figs. 2 and 3 of ref. 14. Two curves of L_1 and L_2 are thus calculated respectively for 122 and 80 μ dia. particles.

To compute the mass transfer term analytically, a mean bubble diameter d_b is needed for each U_f . This size has been given by Morooka¹⁷⁾ for fluid beds with 79 and 195 mm dia. which fluidize 59 and 80 μ dia. FCC catalyst particles. For a 50 mm dia. bed, Furusaki⁵⁾ has measured the number mean bubble size (1.7~4.6 cm for $U_f=10\sim40$ cm/sec) for FCC catalyst of similar size as above. Morooka's number mean bubble size is in 2.0~4.0 cm for $U_f=10\sim40$ cm/sec, and is about the same as that given by Furusaki. Morooka¹⁷⁾ also gives the volume-surface mean bubble size for the same data as above, and this will be taken as the mean bubble diameter d_b for the following computation ($d_b=4.0\sim8.0$ cm for $U_f=10\sim40$ cm/sec and $D_T=79$ and 195 mm).

Assuming freely rising bubbles through the dense phase, $k_{ob}a_b/\varepsilon_b$ was calculated (refer to Appendix), and is shown in Fig. 2. As has been discussed in Appendix (Section A.2), it seems reasonable to take into consideration the influence of the Hatta number β_r on k_{ob} . This influence is also observed from the predicted curves of A and B as shown in Fig. 2, where larger $K(=\lambda k_r)$ value gives higher $k_{ob}a_b/\varepsilon_b$. Due to this, Eq. (A.6) has been utilized to predict the rate of gas exchange during reaction.

In Fig. 2, the cross-hatched curve A has been determined from Eqs. (A.6)~(A.9), and Curve B is from the same equations by utilizing Eq. (A.15) instead of Eq. (A.7). Prediction seems to be about reasonable. Curve A, which has been calculated from the bubble sizes originally determined for 59 and 80 μ dia. FCC particle beds, fits fairly well with the data for 80 μ dia. alumina particle bed. This curve, however, fails to account for the data for 122 μ dia. particle bed. Also, visually it is somewhat difficult to believe that bubble sizes larger than the bed diameter of 5 cm exist in the bed. Perhaps the effective bubble sizes may be somewhat smaller than those assumed here, and the increase in $k_{ob}a_b/\varepsilon_b$ due to decrease in the bubble sizes is cancelled as the result of the rising velocity of bubbles being retarded by the wall effect^{13, 21, 22)}. More work is needed to make these phenomena clear.

4. Test for Data by Gilliland and Knudsen⁷⁾

* $\varepsilon_b=0.12, 0.18, 0.23, 0.27$, and 0.30 for $U_f=10, 20, 30, 40$, and 50 cm/sec respectively. These ε_b are somewhat lower than those given by Morooka *et al.*¹⁸⁾

** Graphical plotting of $KQ/(K-Q)$ vs. $K^2/(K-Q)$, however, gives good straight lines for each constant U_f series. Another source of inaccuracy is the assumption of the reaction being isothermal even in the dilute phase.

Fig. 2 Comparison of overall mass transfer term $k_{ob}a_b/\varepsilon_b$, as a function of U_f from several sources.

Reaction data have been given by them⁷⁾ for a 2-in. diameter fluidized bed reactor, using ethylene hydrogenation on various particle sizes of copper-impregnated fluid cracking catalyst. Their series 52, 54 and 56 runs furnish enough information to be analysed further by the "direct contact model" (refer to their Figs. 4 and 5). Two terms of F and " a " are obtained by plotting $KQ/(K-Q)$ vs. $K^2/(K-Q)$ in essentially same way as Lewis *et al.*¹²⁾

The mass-transfer term $k_{ob}a_b/\varepsilon_b$ calculated from F , which is obtained as above, is plotted as Curve G in Fig. 2 for 120 μ dia. catalyst particles. The curve is located in about the same order of magnitude as those by Lewis.

The fraction of catalyst " a " is also plotted in Fig. 1a as a function of U_f . The catalyst activity K converted from Gilliland's intrinsic activity for those runs is 0.83, 0.48 and 0.24 sec^{-1} for series 52, 54 and 56 respectively, and is fairly lower than those by Lewis ($K=1.29\sim14.3 \text{ sec}^{-1}$). Due to this, accuracy for determining F and " a " may not** be so good as that in the case of Lewis' data. With this in mind, it is still quite interesting to see that the term " a " is surprisingly greater than those by Lewis.

When " a " calculated as above is converted to μ^* by utilizing Eq. (2), μ^* amounts to 7.6 to 5.5 for $U_f=10\sim50$ cm/sec. In other words, the volume of catalyst (with an apparent density the same as the emulsion phase) which contacts freely with the bubble gas amounts to 7.6 to 5.5 times the volume of the bubble gas itself. Obviously this is not sound physically, even if the above μ^* are rather overestimated due to lack of accuracy**.

On the other hand, this amount of freely contacting catalyst could possibly be suspended in the dilute phase, since the catalyst had a much less quiescent bed density of 0.33 g/cc. There is an experimental observation which supports this possibility. Utilizing the fluid bed shown in Fig. 1 of ref. 14, bed density distribution was measured for air-fluidized microspherical alumina particles (MSC-1A pre-

pared by Mizusawa Kagaku Co.) with 88 μ average diameter. The quiescent bed density was 0.833 g/cc. L_q was changed from 20 to 60 cm, and U_f from 20 to 60 cm/sec. The volume fraction of particles suspended in the dilute phase decreased for MSC-1A particles to about one half of that for FCC catalyst of 85 μ diameter (ref. to Fig. 4 of ref. 14). Thus, at least qualitatively, it is quite probable for the series by Gilliland that a fair amount of catalyst particles is suspended in the dilute phase.

5. Discussion and Remarks

The kinetic approach presented here as the "successive contact model" has been formulated in its simplest version for more complicated fluid bed behaviour (Sections 2.2 and 2.3 of ref. 14). In spite of this, the test of the model has not revealed any contradiction with observations (Sections 3 and 4). Quite large μ^* obtained in Section 4 seems to provide support to this approach. Physical quantities introduced into this new model are measurable by independent experimental methods, as have been estimated frequently in Section 2~4.

In fluid beds without internals, progress of reaction in the dilute phase has been revealed as having an important role in overall conversion under relatively high gas velocity. The reaction in the dilute phase seems to proceed under relatively low axial dispersion of reacting gas. When the reaction proceeds extensively in the phase, a temperature distribution is built up axially. This distribution has no particular ill effect on the progress of simple irreversible reaction, but may reveal sometimes a drastic effect on the selectivity of successive or simultaneous reaction. The selectivity depends on the relative magnitude of activation energies of each reaction step, and is influenced directly by the temperature distribution.

Hence, when the temperature does not too much influence the selectivity, and the rate of reaction is enough high, progress of reaction in the dilute phase would even be desirable, because good fluid-particle contact is realized in the dilute phase under higher gas velocity.

Fluid beds with internals are frequently utilized for exothermic reactions. Qualitative implication of the internals has been discussed extensively^{8,26)}. Lewis¹²⁾ have shown quantitatively the influence of baffles on the progress of reaction. Overall effect of the internals is, thus, supposed first to increase $k_{ob}a_b$ and perhaps μ (not μ^*), and secondly to decrease the catalyst content in the dilute phase by reducing both the circulation rate of the dense phase and the ascending velocity of bubbles.

The successive contact model seems to provide a reasonable approach to fluid bed design, by making

it feasible to take into consideration the influence of bed design as well as the scheme and rate of a given reaction on the optimum selectivity for overall progress of the reaction.

Conclusion

1. An apparent fraction of catalyst in the bubble phase, μ^* , for catalytic hydrogenation of ethylene by fluid beds is shown to be reasonably well estimated from catalyst distribution in the dilute phase. The new approach provides a sound explanation for an apparent anomaly of μ^* being fairly greater than unity for some published data.
2. Influence of bed design and operation on selectivity of a given reaction is briefly given for fluid beds.
3. The concept of diffusion with simultaneous adsorption and chemical reaction is shown to provide a sound approach for determining the overall coefficient of mass transfer between the bubble void and the emulsion phase during reaction in a fluid bed.

Appendix

A-1 Prediction of gas exchange coefficient

Davidson and Harrison³⁾ have assumed that there are both convective and diffusional flows which contribute to the gas exchange between the bubble and emulsion phases. Partridge and Rowe²⁰⁾ have developed a boundary layer equation for the transfer from cloud to emulsion phase. Kunii and Levenspiel^{9,10)} have proposed a model for estimating the gas exchange coefficient. They assume that there are two transfer steps, namely the transfer between bubble void and cloud-particle overlap region and that between the cloud-particle overlap region and the emulsion phase. The mass transfer coefficient for the former step is assumed as being given by that of Davidson and Harrison, and for the latter step the Higbie penetration model is supposed to be well applicable. In terms of mass transfer coefficient their equation is given as follows:

$$1/k_{ob} = 1/k_{bc} + 1/k_{ce} \quad (\text{A.1})$$

where

$$k_{bc} = (3/4)U_{mf} + 0.975D_G^{1/2}(g/d_b)^{1/4} \quad (\text{A.2})$$

$$k_{ce} \doteq (2/\sqrt{\pi})(\varepsilon_{mf}^2 D_G u_b / d_b)^{1/2} \quad (\text{A.3})$$

Recently, Chiba and Kobayashi¹⁾ have presented the following equation based on the Murray stream function¹⁹⁾:

$$k_{ob} = \frac{2}{\sqrt{\pi}} \left(\frac{\varepsilon_{mf}^2 D_G u_b}{d_b} \right)^{1/2} \left(1 - \frac{1}{\alpha} \right)^{1/2} \quad (\text{A.4})$$

They further introduce the influence of physical adsorption equilibrium on particles. Their relation is modified as follows in terms of present notation, by using the fact that ε_{mf} is nearly the same as ε_{fe} :

$$(k_{ob})_{m_s \neq 0} / (k_{ob})_{m_s = 0} = \eta \sqrt{m / \varepsilon_{fe}} \quad (\text{A.5})$$

where

$$\eta = \sqrt{1 + \frac{2}{3} \left(1 - \frac{\varepsilon_{fe}}{m} \right) \left(\frac{1}{\alpha - 1} \right)}$$

Gas exchange during catalytic reaction between bubbles and the emulsion phase of catalyst particles is essentially a diffusion phenomenon with simultaneous adsorption and chemical reaction in the emulsion phase. Based on this physical

picture, Miyauchi and Morooka¹⁶⁾ have developed the following relation for k_{ob} .

$$1/k_{ob} = 1/k_b + 1/\beta_r k_e \quad (\text{A.6})$$

where

$$k_b = (2/\sqrt{\pi}) \sqrt{D_G u_b/d_b} \quad (\text{A.7})$$

$$k_e = (2/\sqrt{\pi}) \sqrt{m D_{eff} u_b/d_b} \quad (\text{A.8})$$

and

$$\beta_r = \beta - (\varepsilon_{fe}/m)J \quad (\text{A.9})$$

The term β is the Hatta number* for unsteady gas absorption with chemical reaction as given by Danckwerts²¹⁾, and is found in standard texts as a function of $m_H (= \sqrt{k_r D_{eff}}/k_e)$. J is the term associated with the gas cloud, and is found from Fig. 3 as a function** of m_H and Pe (the gas cloud parameter).

Relation between the proposed equations is as follows. Eq. (A.6) is about the same as Eq. (A.1) for smaller U_{mf} , when $\beta_r = 1$ (no reaction) and $m = \varepsilon_{mf}$ (no partition to particles). The latter condition is that Eqs. (A.3) and (A.8) are different in general, due to m being not necessarily equal to ε_{mf} . Bubble void resistance to mass transfer has been assumed as being negligible in Eq. (A.4). This equation is applicable to the case of arbitrary m by utilizing Eq. (A.5). Eq. (A.8) is rewritten in similar form to Eq. (A.5) as follows (note that $m = \varepsilon_{fe} + m_s \varepsilon_{se}$, and $\varepsilon_{fe} \doteq \varepsilon_{mf}$):

$$(k_e)_{m_s \neq 0}/(k_e)_{m_s = 0} = \sqrt{m/\varepsilon_{fe}} \quad (\text{A.10})$$

The correction factor η to Eq. (A.5) is nearly equal to unity for $\alpha \geq 10$, so that Eq. (A.5) is almost equivalent to Eq. (A.10) for smaller particles. Eq. (A.4) is independent of m_H or the rate of chemical reaction.

Recently, Drinkenburg and Rietema⁴⁾ have presented a numerical computation of k_{ob} based on the stream functions given by Davidson and Harrison³⁾ and by Murray¹⁹⁾. The bubble void resistance to mass transfer has been neglected. Enhancement of gas transfer rate by diffusion with simultaneous chemical reaction (Fig. 5 of ref. 4) seems to be reasonably well expressed by Eq. (A.9), when the enhancement is expressed as the Hatta number. Enhancement by physical adsorption (Fig. 2 of ref. 4) is also approximated by Eq. (A.5) or Eq. (A.10) for smaller particles.

When the reaction data by Lewis¹²⁾ have been analysed by utilizing Eq. (A.6), it has turned out that β_r amounts to 1.45 (free flow bubbles) and higher values than 1.45 (bubbles retarded by the wall effect) for the highest reaction rate constant of $K = \lambda k_r = 14.3 \text{ sec}^{-1}$. Hence it seems reasonable to take into consideration the influence of β_r on k_{ob} . Also, it has been shown experimentally that k_b is not necessarily negligible for fluid beds²³⁾.

Due to these, Eq. (A.6) has been utilized, in what follows, to predict the rate of gas exchange during reaction.

When $Pe \leq 10^{-2}$ and $m_H \geq 3$, Eq. (A.9) reduces to $\beta_r \doteq \beta \doteq m_H (= \sqrt{k_r D_{eff}}/k_e)$. Hence, the following simple relation is obtained for k_{ob} from Eq. (A.6).

$$* \beta = \left(m_H + \frac{\pi}{8m_H} \right) \operatorname{erf} \left(\frac{2m_H}{\sqrt{\pi}} \right) + \frac{1}{2} \exp \left(-\frac{4m_H^2}{\pi} \right) \quad (\text{A.11})$$

$$** J = \frac{\pi}{4m_H} \operatorname{erf} \left(\frac{2m_H}{\sqrt{\pi}} \right) - \int_0^1 \frac{1}{2\sqrt{\phi}} \exp \left[-\left(\frac{4m_H^2}{\pi} \phi + \frac{Pe}{\phi} \right) \right] d\phi \quad (\text{A.12})$$

where ϕ is an integration parameter, and

$$(J)_{m_H=0} = 1 - e^{-Pe} + \sqrt{\pi Pe} \operatorname{erfc} \sqrt{Pe}; (J)_{m_H \gg 1} = 0,$$

$$(J)_{m_H=0, Pe \leq 0.1} = \sqrt{\pi Pe} - Pe, (\text{max. error} = -2.6\%)$$

and

$$(J)_{Pe=0} = 0.$$

Fig. 3 Integral J as a function of Pe and m_H

$$1/k_{ob} = 1/k_b + 1/\sqrt{k_r D_{eff}} \quad (\text{A.13})$$

k_{ob} is independent of k_e under such circumstances.

A-2 Computation of k_{ob} by utilizing Eq. (A.6)

Physical data necessary for the computation are D_G , m , k_r , U_{mf} , d_b and ε_{fe} ($\doteq \varepsilon_{mf} \doteq 0.50$).

By knowing these basic data, several quantities are then computed from the following relations:

$$u_b = 0.71 \sqrt{g d_b} \text{ (free flow bubble)}^{3)}$$

$$\alpha = u_b/(U_{mf}/\varepsilon_{mf})$$

$$\delta = \frac{d_b}{2} \left[\left(\frac{\alpha}{\alpha-1} \right)^{1/3} - 1 \right]; \text{ (ref. 19)}$$

$$D_{eff} = \varepsilon_{fe} D_G / \chi \doteq D_G / 3.0 \quad (\because \varepsilon_{fe} \doteq 0.5, \chi \doteq 1.5)$$

$$k_b = \text{Eq. (A.7)}, \quad k_e = \text{Eq. (A.8)}$$

Since the particle Péclet number $d_p U_{mf}/D_G$ for the emulsion phase in fluid beds is usually below 0.1 (refer to the footnote of Section 3.2 of ref. 14), the effective diffusivity may be well expressed by the above equation^{6,15)}, where the tortuosity factor is taken according to the value given by Hoogschagen¹⁵⁾.

Pe and m_H are then obtained as follows:

$$Pe = m^2 \delta^2 u_b / (4 d_b D_{eff})$$

$$m_H = \sqrt{k_r D_{eff}} / k_e$$

By knowing m_H , β in Eq. (A.9) is determined from the Hatta number for gas absorption with chemical reaction. Also, J is obtained from Fig. 3 for most fluid-bed operations. Hence β_r is calculated from Eq. (A.9), and k_{ob} from Eq. (A.6).

The computation procedures are considerably simplified when $Pe \leq 0.005$. k_{ob} is simply given by

$$1/k_{ob} = 1/k_b + 1/\beta k_e \quad (\text{A.14})$$

According to Morooka¹⁷⁾, the volume-surface mean diameter of bubbles, d_b , is approximately given as 4.0, 5.5, 7.5, 8.0 and 8.0 cm for $U_f = 10, 20, 30, 40$ and 50 cm/sec , respectively.

Assuming that these bubble diameters are also applicable to 122μ dia. FCC catalyst and 80μ dia. alumina catalyst, both utilized by Lewis¹²⁾, k_{ob} are calculated by Eq. (A.6), where $m = 1$ and $D_G = 0.904 \text{ cm}^2/\text{sec}$ for $\text{C}_2\text{H}_4\text{-H}_2$ system at 113°C and 1 atm . Also, $U_{mf} = 0.73 \text{ cm/sec}$, $\varepsilon_{fe} = 0.492$ (average), and $k_r = K/1.09$ with $K = 1.29 \sim 14.3 \text{ sec}^{-1}$ according to Lewis¹²⁾. $k_{ob} a_b / \varepsilon_b (= 6 k_{ob} / d_b)$ thus obtained is shown in Fig. 2 as the cross-hatched curve A.

Experimentally measured bubble-phase coefficient of mass

transfer has been given by Yamazaki²³⁾ as

$$k_b a_b / \varepsilon_b (= 6k_b / d_b) = 3.66 U_f^{1/3} (0.66 / Sc)^{1/2} \quad (\text{A15})$$

for FCC catalyst particles of 59 μ dia. fluidized by air in 79 and 195 mm dia. columns. By using this k_b (instead of Eq. (A.7)) for Eq. (A.6), $k_{ob} a_b / \varepsilon_b$ is also calculated and illustrated as Curve B in Fig. 2, where $Sc = 0.167$ for C_2H_4 - H_2 system.

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Nomenclature

(See Ref. 14 for additional nomenclature)

D_T	= bed diameter	[cm]
D_{eff}	= effective diffusivity	[cm ² /sec]
F	= ref. to Eq. (3)	[1/sec]
H	= manometer reading	[cm]
$\bar{H}_0, H_f, H_t$	= H at $z=0, L_f$ and L_t respectively	[cm]
J	= refer to Eq. (A.12)	[—]
K	= catalyst activity, Eq. (1)	[1/sec]
Pe	= $m\delta^2 u_b / 4d_b D_{eff}$	[—]
Q	= specific converting power	[1/sec]
Sc	= Schmidt number	[—]
a	= refer to Eq. (2)	[—]
d_b	= bubble diameter	[cm]
k_b, k_e	= film coefficients for bubble phase and emulsion phase, respectively	[cm/sec]
k_{be}, k_{ee}	= refer to Eqs. (A.2) and (A.3)	[cm/sec]
m_H	= $\sqrt{k_r D_{eff}} / k_e$	[—]
u_b	= mean interstitial velocity of bubbles relative to the emulsion phase	[cm/sec]
α	= ratio of u_b to $U_{mf} / \varepsilon_{mf}$	[—]
β	= Hatta number for gas absorption with reaction, Eq. (A.11)	[—]
β_r	= refer to Eq. (A.9)	[—]
δ	= thickness of cloud-particle overlap region	[cm]
ε_{mf}	= void fraction at U_{mf}	[—]
η	= refer to Eq. (A.5)	[—]

λ	= $\varepsilon_{sq} / \varepsilon_{se} \doteq 1.09$	[—]
χ	= tortuosity factor $\doteq 1.5$	[—]

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