Relative Contribution of Variables Affecting the Reaction in Fluid Bed Contactors

Simplified expressions to calculate the extent of reaction in a fluid bed show good agreement with the experimental data of Lewis et al. (1959) and Swaay and Zuiderweg (1972). The contribution of the dilute phase seems more important than that of the jetting zone or reaction factor (Hatta number) in usual fluid bed operations. The effect of the dilute phase is discussed. It is profitable to use the dilute phase for the consecutive isothermal and endothermic reaction, but unprofitable for the exothermic reaction. These characteristics depend upon the nature of reactions and careful considerations as to the choice of the gas velocity, particle characteristics, and reactor design will be necessary.

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SCOPE

Extensive studies on fluidized bed reactors have been made and many reactor models have been proposed (Shen and Johnston, 1955; Mathis and Watson, 1956; Lewis et al., 1959; Orcutt et al., 1962; Partridge and Rowe, 1966; Kobayashi et al., 1969; Kunii and Levenspiel, 1969; Kato and Wen, 1969). Essentially, these models are variations of the two-phase concept (Shen and Johnston, 1955). Some take into consideration the gas cloud region associated with each bubble. The main differences between the models are related to whether or not some fraction of the catalyst is in direct contact with the bubble gas, the extent of axial mixing in each phase, and axial change in bubble size. Catalytic reactions were reported with respect to these models, but data for fast reactions are especially important in studying the effect of mass transfer in the reactor. Miyauchi and Morooka (1969b) analyzed the data of Lewis et al. (1959) on the hydrogenation of ethylene and found that the estimated bubble size became smaller for fast reactions. This phenomenon was discussed by Gilliland and Knudsen (1971) and also found by Swaay and Zuiderweg (1972) in the decomposition of ozone and by Furusaki (1973) in the oxidation of hydrogen chloride. They found the apparent mass transfer rate between the bubble and the emulsion became greater for the case of high reaction rates. Miyauchi (1974b) attributed this to the effect of catalysts suspended in the dilute phase. He considers the role of catalysts in the dilute phase in his successive contact model instead of assuming the catalyst in the bubble phase (Mathis and Watson, 1956; Lewis et al., 1959). In this model, the dense phase consists of the bubble and emulsion phases with cloud region between them. The dilute phase is located above the dense phase. Reaction proceeds successively first in the dense phase and next in the dilute phase.

Taking account of the effect of catalysts in the dilute phase, it is important to consider the effect of temperature distribution in the dilute phase since energy transport in this phase is considerably smaller compared with that in the dense phase. The heat capacity in the dilute phase is also less. Homogeneity of temperature cannot be achieved in the dilute phase. This phenomenon is expected to cause a different selectivity of the reaction.

This article presents further developments of the model and gives a simple form for HRU and contact efficiency in order to be able to be useful for design. Using the model, the effect of the dilute phase is compared with other factors such as the contribution of the reaction factor due to simultaneous reaction and mass transfer between bubble and emulsion and the contribution of the jet above the distributor. Also, it presents the effect of the dilute phase on the selectivity of isothermal and nonisothermal reactions for several reaction systems. The industrial importance of the control of the dilute phase is discussed.

CONCLUSIONS AND SIGNIFICANCE

After several simplifications applicable to the usual fluid bed operations, the expressions for the overall height of a reaction unit and the contact efficiency are presented in this paper. The calculations give the relative contribution of variables such as the modified Hatta number β_r, the better contact near the distributor, and the effect of catalyst particles suspended in the dilute phase. Of these, the last is found as a major cause of the significant behavior for the case of large reaction rates. The former two factors are less important for most of the fluid bed. The contribution of the dilute phase becomes significant for the case of first order reaction rate constant k_r greater than $1 \sim 5 s^{-1}$. The contribution of β_r becomes significant for k_r greater than $20 \sim 30 s^{-1}$.

The temperature effect in the dilute phase is an important factor in controlling the selectivity of reactions. Whether it is profitable to enlarge the extent of reaction in the dilute phase depends upon the mechanism of the reaction. In case of the reaction $A \rightarrow B \rightarrow C$ in which B is the required product, it is more profitable to use the dilute phase for endothermic reactions, but it is advisable to finish the reaction in the dense phase for exothermic reactions. For the reaction $\begin{array}{l} A \rightarrow X \rightarrow Y \\ \quad \searrow P \ \searrow Q \end{array}$ in which Y is the desired product, it is desirable to use the dilute phase. However, for the reaction, $\begin{array}{l} A \rightleftarrows X \\ \quad \searrow Y \end{array}$ in which X is the desired product, it is desirable to finish the reaction in the dense phase.

SIMPLIFIED TREATMENT OF REACTIONS IN A FLUID BED

Mass Transfer Coefficients

Since a fluid bed is a heterogeneous reactor, mass transfer between the bubble and emulsion phases is the most important factor affecting the efficiency in the dense phase. The overall mass transfer coefficient k_{ob} has been proposed by several investigators (Davidson and Harrison, 1963; Partridge and Rowe, 1966; Kunii and Levenspiel, 1968; Miyauchi and Morooka, 1969a; Chiba and Kobayashi, 1970; Drinkenburg and Rietema, 1972; Miyauchi, 1974b). Considering these theories and the conditions accompanied by fluid beds, one can generalize the mass transfer coefficient with the following assumptions: (1) the reactant is in adsorption equilibrium with the catalyst particles (Tam and Miyauchi, 1971). (2) The thickness of cloud is small. (3) Reaction is considered to be the first order and irreversible. (4) Reactant penetration through the emulsion phase is a diffusion phenomenon with simultaneous adsorption and reaction. Then, k_{ob} can be calculated in a following way for usual fluid bed operations ($\alpha \gtrsim 10$):

$$\frac{1}{k_{ob}} = \frac{1}{k_b} + \frac{1}{\beta_r k_e} \tag{1}$$

$$\beta_r = \beta - \left(\frac{\epsilon_{fe}}{m}\right) J \tag{2}$$

k_b and k_e can be calculated by any theory such as Kunii and Levenspiel when the effect of adsorption is absent, but it is normally better to use experimental values. If the mass transfer process obeys the penetration theory, they are calculated by Equations (3) and (4) with $\tau_b = d_b/u_b$, where $u_b = 0.711\sqrt{gd_b}$. β is the usual reaction factor (Hatta number) for unsteady gas absorption with the first-order irreversible chemical reaction (Danckwerts, 1950). J is a term associated with the gas cloud and is found from Equation (5). (See Figure 3 of Miyauchi, 1974b).

$$k_b = \frac{2}{\sqrt{\pi}}\sqrt{\frac{D_G}{\tau_b}} \tag{3}$$

$$k_e = \frac{2}{\sqrt{\pi}}\sqrt{\frac{m\,D_{\text{eff}}}{\tau_b}} \tag{4}$$

$$J = \frac{\pi}{4m_H}\,\text{erf}\left(\frac{2m_H}{\sqrt{\pi}}\right) - \int_0^1 \frac{1}{2\sqrt{x}}\exp\left[-\left(\frac{4m_H{}^2}{\pi}x + \frac{Pe}{x}\right)\right]dx \tag{5}$$

Physical data necessary to compute k_{ob} are D_G, m, k_r, u_{mf}, d_b, and ϵ_{fe}. ϵ_{fe} can be approximated by ϵ_{mf}. With Higbie's penetration theory, k_{ob} can be expressed by Equation (6).

$$k_{ob} = \frac{2/\sqrt{\pi}}{1 + \dfrac{1}{\beta_r}\sqrt{\dfrac{\chi}{m\,\epsilon_{fe}}}}\sqrt{\frac{D_G u_b}{d_b}} \tag{6}$$

where the tortuosity factor χ can be approximated by 1.5 (Miyauchi, 1971).

Simplified Modeling of a Fluid Bed

General features of flow and reaction in a fluid bed have been given by Squires (1962) and Ikeda (1963). The bed consists of the dense phase and the dilute phase; the emulsion and bubble phases constitute the former. The emulsion phase reveals liquid-like properties during fluidization.

Intense circulation of the dense phase results from the buoyant force induced between the centrally ascending bubble-rich phase and the peripherally descending emulsion phase of low bubble content (Lewis et al., 1962; Morooka et al., 1971). Measurements under relatively high gas velocity of practical interest ($U_f \gtrsim 10$ cm/s) show that the rate of circulation is higher than that due to solid particles conveyed by the bubble wake. The behaviour resembles, in many respects, that in gas bubble columns (Towell et al., 1965). Thus, volumetrically half of the emulsion phase is in the up-flow zone and another half is in the down-flow zone.

In contrast to the teeter beds (Squires, 1962), the mean bubble size in a fluid bed is almost unchanged with axial bed height (Morooka et al., 1971). The size seems to be characteristic of the flow intensity in the bed. Thus, the general flow feature of the fluid bed operated under relatively high gas velocity may briefly be sketched as illustrated in Figure 1.

Also, the following simplifying assumptions are taken for the dense phase: 1. Constant ascending and descending velocities for the bubble phase and the circulating emulsion phase, 2. negligible influence of axial dispersion of the emulsion phase on the progress of reaction, 3. the bubble phase is included only in the ascending dense phase, and 4. U_{mf} is neglected in comparison with U_f for the fluid bed operated under relatively high gas velocity.

Besides the above, another simplification of the usual fluid bed reactor operation is to place a restriction on φ, that

$$\varphi = \frac{U_f}{mU_e}\left(1 + 4m\frac{k_{ex}a_{ex}}{k_{ob}a_b} + \frac{\epsilon_e\,k_r}{k_{ob}a_b}\right) \gtrsim 10 \tag{7}$$

where U_e is the superficial circulation velocity of the emulsion phase.

This restriction has been derived by solving the differen-

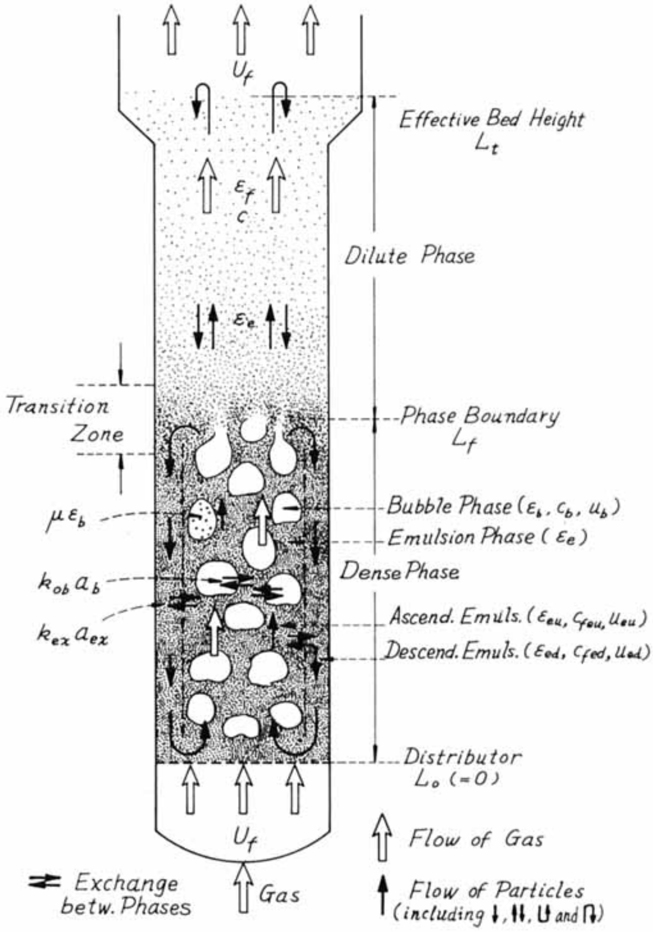

Fig. 1. Reactor model for a fluid bed.

tial equations which govern the reactor model given in Figure 1 and is a measure of the influence of radial mixing on the overall progress of the reaction (Miyauchi and Morooka, 1969b). The influence of the emulsion phase circulation on the extent of reaction apparently disappears when φ satisfies this restriction.

In small scale fluid beds under relatively high gas velocities, $k_{ex}a_{ex}/k_{ob}a_b$ is usually much greater than unity. The restriction is satisfied under such circumstances.

Overall Conversion of a Fluid Bed

With the restriction [Equation (7)] for φ, the emulsion phase circulation can be neglected, and the following material balance equations are obtained in dimensionless forms for the dense phase:

$$-\frac{dC_b}{dZ} - N_{ob}(C_b - C_{feu}) - \mu\epsilon_b N_r C_b = 0 \qquad (8)$$

$$N_{ob}(C_b - C_{feu}) - N_{ex}(C_{feu} - C_{fed}) - (\epsilon_e N_r/2)C_{feu} = 0 \qquad (9)$$

$$N_{ex}(C_{feu} - C_{fed}) - (\epsilon_e N_r/2)C_{fed} = 0 \qquad (10)$$

where half of the emulsion phase is in the up-flow and another half is in the down-flow. Equation (8) is for the bubble phase. Equations (9) and (10) are for the ascending and descending emulsion phases, respectively. If the following nondimensional parameters are defined from Equations (9) and (10), one can simplify Equation (8) into Equation (13). ξ is the fraction of catalyst in the dense phase which is utilized effectively for the reaction.

$$1/N_{or} = 1/N_{ob} + 1/(\xi\,\epsilon_e N_r) \qquad (11)$$

$$\xi = 1/2 + 1/(2 + \epsilon_e N_r/N_{ex}) \qquad (12)$$

$$-\,dC_b/dZ = (N_{or} + \mu\,\epsilon_b N_r)C_b \qquad (13)$$

For the dilute phase, the diffusion model approach for axial mixing may be more general (Handlos et al., 1957). However, the usual piston flow assumption is taken here for simplicity, since catalyst particles in the dilute phase are seemingly suspended rather uniformly in small scale fluid beds. Thus, one has

$$-\,dC/dZ - \epsilon_e N_r C = 0 \qquad (14)$$

where $N_r = k_r L_f/U_f$. Integrating Equations (13) and (14), the overall conversion in the whole bed is simply given by

$$C_{fo}/C_f^0 = \exp\{-[N_{or} + (\mu\epsilon_b + e)N_r]\} \qquad (15)$$

where $e = \int_1^{Z_t} \epsilon_e dZ$. e, in terms of the volume fraction of emulsion, is the average of the volume fraction occupied by catalyst particles in the dilute phase. This value is to be calculated from density distribution measurements in the dilute phase.

Expression of the Bed Behavior by HRU

The relative contribution of the variables on the overall performance of a fluid bed is more clearly visualized by applying the concept of the overall height of a reaction unit H_{oR} (Hurt, 1943) for a first-order reaction. H_{oR} is defined by

$$C_{fo}/C_f^0 = \exp(-N_{oR}) = \exp(-L_f/H_{oR}) \qquad (16)$$

where N_{oR} is the overall number of a reaction unit. Equations (15) and (16) lead to the following relation:

$$L_f/H_{oR} = N_{oR} = N_{or} + (\mu\epsilon_b + e)N_r \qquad (17)$$

As defined in the Notation, $H_b = U_f/k_b a$, $H_e = U_f/k_e a$, $H_{ob} = U_f/k_{ob}a_b$, and $H_r = U_f/k_r$. Also, Equation (11) indicates that H_{or} ($= L_f/N_{or}$) equals $H_{ob} + H_r/\xi\epsilon_e$. Hence, H_{oR} is given as follows:

$$1/H_{oR} = 1/(H_{ob} + H_r/\xi\epsilon_e) + (\mu\epsilon_b + e)/H_r \qquad (18)$$

where $H_{ob} = H_b + H_e/\beta_r$ from the use of Equation (1). Equation (18) is rewritten in terms of usual physical quantities as follows:

$$H_{oR} = \frac{U_f}{\left(\dfrac{1}{1/k_{ob}a_b + 1/\xi\epsilon_e k_r}\right) + (\mu\epsilon_b + e)k_r} \qquad (19)$$

For later use it is convenient to define the overall rate coefficient of reaction k_{oR} as follows:

$$H_{oR} = U_f/k_{oR} = L_f/N_{oR} \qquad (20)$$

Expression for the Contact Efficiency of the Whole Bed

The expression for H_{oR} can be modified to express the contact efficiency η_r. If one defines η_r as $N_{oR}/(N_{oR})_{\rm FIX}$, where $(N_{oR})_{\rm FIX}$ is the number of a reaction unit for a piston flow packed bed reactor, the overall extent of reaction for a fluid bed is given by

$$\frac{C_{fo}}{C_f^0} = \exp[-\eta_r(N_{0R})_{\rm FIX}] \qquad (21)$$

This definition of η_r is used by Lewis et al. (1959) and Gilliland and Knudsen (1971) as E ($= Q/K$). If the total amount of catalyst in a fluid bed (expressed as emulsion) is given by $L_f(\epsilon_e + \mu\epsilon_b + e) = L_f\epsilon_e^*$, the reaction rate constant for the packed bed is given by

$$K = \frac{L_f\epsilon_e^*}{L_q}k_r$$

since then,

$$\eta_r = \frac{k_{oR}L_f}{U_f}\cdot\frac{U_f}{KL_q} = \frac{1}{\epsilon_e^*}\cdot\frac{k_{oR}}{k_r}$$

From Equation (20) for k_{oR} and Equation (19) for H_{oR},

$$\eta_r = \frac{1}{\epsilon_e^*}\left[\frac{1}{k_r/k_{ob}a_b + 1/\xi\epsilon_e} + (\mu\epsilon_b + e)\right] \qquad (22)$$

This equation allows determination of the contact efficiency of the whole bed. The effective reaction rate in a fluid bed is η_r times the reaction rate in a packed bed. Special cases arise for small or large reaction rates.

$$\lim_{k_r << k_{ob}a_b} \eta_r = 1 \qquad (23)$$

$$\lim_{k_r >> k_{ob}a_b} \eta_r = \frac{1}{\epsilon_e^*}(\mu\epsilon_b + e) \qquad (24)$$

For a fluid bed, Equation (24) is approximated by

$$\lim_{k_r >> k_{ob}a_b} \eta_r \doteq \frac{e}{\epsilon_e^*} \qquad (25)$$

since the density of catalyst particles in the bubble phase is much less than that in the dilute phase.

Effect of the Longitudinal Bubble Size Distribution

Several investigators consider the bubble size to increase with the bed height (Kato and Wen, 1969; Mori and Muchi, 1972; Chiba et al., 1973). However, for a fluid bed, in which the average particle size is usually less than 150μ (Squire, 1962) and U_f/U_{mf} is more than 30, the following observations are shown by Morooka et al. (1971): (1) Bubbles from a single nozzle break up in rising a certain distance to attain a final size. (2) Bubbles from a perforated plate associate together when rising. (3) d_b stays fairly constant longitudinally. Basov et al. (1969) mea-

TABLE 1. EFFECT OF THE DISTRIBUTION OF $k_{ob}a_b$ TO $\overline{k}_{or}$

k_r	0.1	0.2	0.5	1	5	10	50
$\overline{k}_{or}$	0.0631	0.0947	0.141	0.184	0.221	0.230	0.237
$k_{or})_{\text{app.}}$	0.0705	0.1089	0.162	0.193	0.228	0.233	0.238
Error, %	11.7	15.0	14.9	4.9	3.2	1.3	0.5

sured the longitudinal density distribution of particles by a radiation densimeter for crushed SiO_2-Al_2O_3 cracking catalyst ($d_{p\text{ Ave.}} = 120 \sim 130\mu$). From their observations, it is seen that near the distributor ($0 \sim 20$ cm above) there is a region in which the density varies with height and then it stays constant. Circulation in the dense phase is low near the distributor. Also, they observed that the density of particles is low in that region since the particles are not supplied from the emulsion above.

From these results, one may consider the bottom region of a fluid bed to be different from those above. This lower region may be called the *jetting zone*. In this zone, d_b may be smaller or larger than the average d_b of the bed, depending upon the character of a distributor and upon the energy input to the jet. Also, circulation is not so extensive that one can neglect the circulating flow of gas and solids in the jetting zone in calculation of conversions. Then, calculations can be accomplished assuming that $k_{ob}a_b$ changes with the distance from the distributor.

With the assumption that $\xi \doteq 1$ (this approximation is reasonable as shown in Figure 4), one obtains the conversion in the dense phase by integrating Equation (13).

$$- \ln \frac{C_{f1}}{C_f^0} = \int_0^1 N_{or} dZ + \int_0^1 \mu\epsilon_b N_r dZ = \overline{N}_{or} + \overline{\mu\epsilon_b N_r} \tag{26}$$

Thus, one may use the average value of N_{or} and $\mu\epsilon_b N_r$ in the calculation. Here $\overline{N}_{or} \doteq N_{or})_{\text{app.}} = 1/[(1/\overline{N}_{ob} + 1/\overline{\xi\epsilon_e N_r})]$ or $\overline{k}_{or} \doteq k_{or})_{\text{app.}} = 1/[(1/\overline{k_{ob}a_b}) + (1/\overline{\xi\epsilon_e k_r})]$. Note that the averaging for $\overline{N}_{or}$ is approximate, but calculations show that the maximum error is about 15%. Table 1 shows the comparison of $\overline{k}_{or}$ for various values of k_r using Equation (27) for the case that $k_{ob}a_b$ is 5 times greater in the first 1/5 part of the bed. $\beta_r \doteq 1$, and $\mu\epsilon_b \doteq 0$ are assumed here. The average $\overline{k_{ob}a_b}$ is taken as 0.239 s^{-1}. This value is an example for the Lewis et al. condition calculated by the penetration theory. (See the next section.) This gives $k_{ob}a_b = 0.665$ s^{-1} for the jetting zone and 0.133 s^{-1} for the other part of the dense phase.

$$\left.\begin{aligned} \overline{k}_{or} &= \int_0^1 \frac{dZ}{\dfrac{1}{k_{ob}a_b} + \dfrac{1}{\epsilon_e k_r}} \\ k_{or})_{\text{app.}} &= \frac{1}{1/\overline{k_{ob}a_b} + 1/\overline{\epsilon_e k_r}} \end{aligned}\right\} \tag{27}$$

Table 1 shows that the maximum error of $\overline{k}_{or}$ neglecting the jetting zone will be about 15% and that the error is smaller for high reaction rates. This last phenomenon clearly indicates that better contact found in fluid beds for high reaction rate systems is not the result of the jet above the distributor. (Of course it is quite possible to have large $k_{ob}a_b$ near the distributor.) The fact that conversions in the fluid bed increase with the amount of catalyst (or the quiescent bed height) also indicates that the contribution of catalyst particles above the jetting zone is considerably large. An exception to this discussion is for fluid beds with fast reactions and with a significant jet above the distributor. In this case, the reaction proceeds extensively in the jetting zone, but catalyst erosion could be excessive. Van Krevelen et al. (1950) showed the behavior of bubbles in liquid as separated bubbles and chain-like bubbles depending upon the Reynolds number of ascending bubbles. Miyauchi and Yokura (1970) gave the relations to obtain bubble diameters issuing from an orifice. In a fluid bed the behavior of the emulsion seems very similar to a liquid. If one considers bubbles from the distributor as the chain-like bubbles and calculate $d_b \doteq \left(\frac{72}{\pi^2} \frac{V_G^2}{g}\right)^{1/5}$, one can show large values ($5 \sim 6$ cm) of d_b for practical values of V_G. Hence, d_b is not necessarily small near the distributor except for one with a homogeneous porous plate under fairly low gas velocity.

Distribution of bubble diameters is sometimes important to calculate $\overline{N}_{ob}$ or $\overline{k_{ob}a_b}$. If k_{ob} is not a function of d_b, the average d_b should be the surface averaged one d_{vs}.

$$d_{vs} = \frac{\Sigma n_i d_{bi}^3}{\Sigma n_i d_{bi}^2} \tag{28}$$

If k_{ob} is considered to be proportional to $d_b^{-1/4}$ from the penetration theory, the average $\overline{d}_b$ should be

$$\overline{d}_b = \left(\frac{\Sigma n_i d_{bi}^3}{\Sigma n_i d_{bi}^{7/4}} \right)^{4/5} \tag{29}$$

Numerically, $\overline{d}_b$ is smaller than d_{vs}, but they are very close together. Sometimes, d_{vs} is considerably larger than the number averaged value (Morooka et al., 1971).

Comparison with the Experimental Data

According to Equation (19), the experimental values of Lewis et al. (1959) and van Swaay and Zuiderweg (1972) are compared with calculated values. In the calculation, k_{ob} is obtained by Equation (6).

D_G is estimated by the method of Hirschfelder et al. (1954). Instead of H_{oR}, van Swaay and Zuiderweg report $(H_{ob})_{\text{app}}$, which is calculated by

$$(H_{ob})_{\text{app}} \equiv \frac{L_f}{(N_{ob})_{\text{app}}} = L_f \left[\frac{1}{N_{oR}} - \frac{1}{\epsilon_e N_r} \right] \tag{30}$$

Comparisons shown in Figures 2 and 3 show good agreement between the experimental data with those calculated. The bed density of van Swaay and Zuiderweg is larger than that of Lewis et al. by the factor of $2.5 \sim 3$. This fact makes one suppose e for the data of van Swaay and Zuiderweg to be about 0.03 at $U_f = 0.15$ m/s. This e is used for our calculations. For the Lewis et al. data, d_b and ϵ_b are measured. e is estimated from a of Lewis et al. and confirmed by the density measurement in the dilute phase (Miyauchi, 1974a; Tsutsui, 1974). Hence, the calculated values have no estimated or adjusted variables.

RELATIVE CONTRIBUTION OF VARIABLES TO THE OVERALL EXTENT OF REACTION

The variables affecting reactions in fluid beds have been discussed in detail. It is important to compare their relative contributions, that is, the modified Hatta number β_r, the jetting zone, and the catalyst dispersed in the dilute

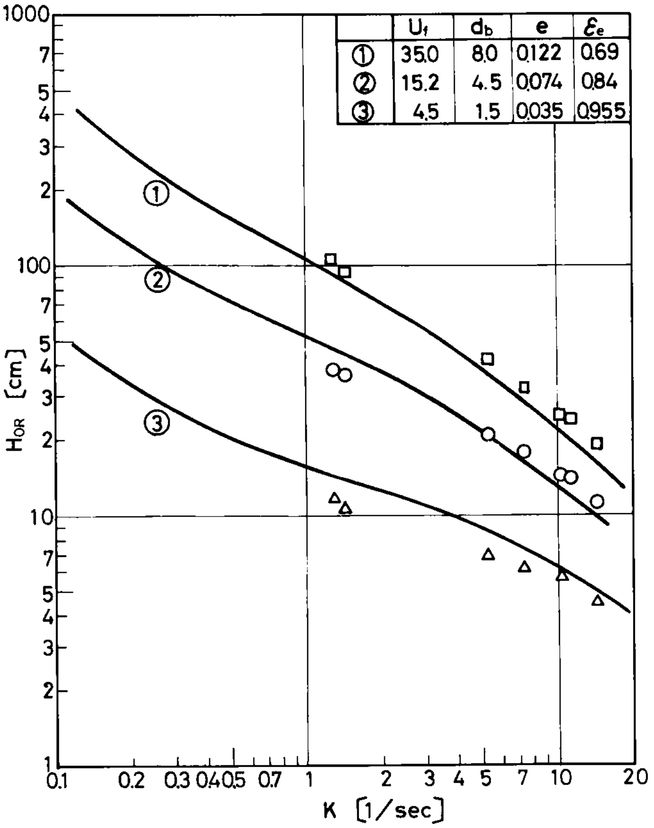

Fig. 2. Comparison of the calculated H_{oR} with the data of Lewis et. al. (1959).

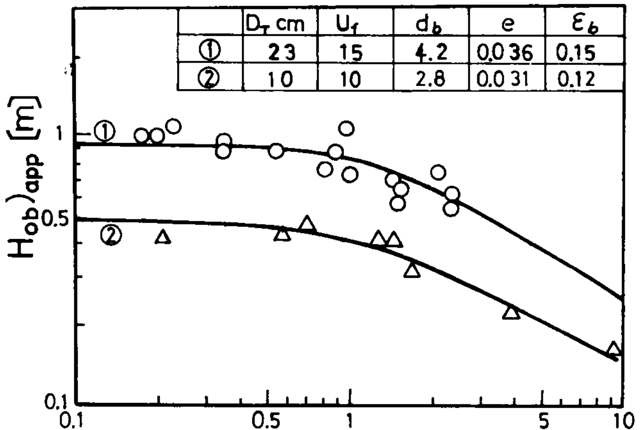

Fig. 3. Comparison of the calculated $(H_{ob})_{\mathrm{app}}$ with the data of van Swaay and Zuiderweg (1972).

phase. For a typical example, take $U_f = 16$ cm/s. In this case, experiments have shown that $\epsilon_b = 0.16$, $d_b = 5$ cm, $\mu \doteq 0$. Also, the catalyst in the bubble in the Lewis et al. analysis is not in the bubble but in the dilute phase. In other words, the fraction of catalyst in the bubble a is equated to $\dfrac{e}{e + \epsilon_e}$. From the Lewis et al. data, for $U_f = 16$ cm/s and $d_p = 122\mu$, $a = 0.081$. e is then calculated as 0.074. For the system C_2H_4-H_2, $D_G = 0.891$ cm^2/s and $D_{\mathrm{eff}} = 0.297$ cm^2/s. Therefore, k_b and k_e are calculated as 3.37 and 1.93 cm/s, respectively.

The values of H_{oR} and η_r are determined for this example by Equations (19) and (22). The results are given in Figures 4 and 5. The effect of ξ is not shown in the figure, but calculations show ξ is approximated by unity for all cases. Also in the figure, the asymptotes of H_{oR} are calculated by

$$\lim_{k_r >> k_{ob}a_b} H_{oR} = \frac{U_f}{e k_r} \tag{31}$$

$$\lim_{k_r << k_{ob}a_b} H_{oR} = \frac{U_f}{\epsilon_e^* k_r} \tag{32}$$

The asymptotes for η_r are given by Equations (23) to (25).

In Figure 4, the broken lines show the results of calculations considering the effect of the jetting zone. From the figures, it is obvious that the effect of e is most significant. β_r is important only for very large values of k_r. In the region of very small k_r, the reaction seems to be the rate determining step. Points where $k_{ob}a_b$, e, and β_r begin to affect H_{oR} are shown in Figure 5 for an approximate estimation. The point for e changed significantly with the amount of catalyst in the dilute phase.

EFFECT OF THE DILUTE PHASE TO THE SELECTIVITY OF COMPLEX REACTIONS

The successive contact model discussed in the previous sections shows the important roles of catalyst dispersed in the dilute phase. It is expected that in case of isothermal

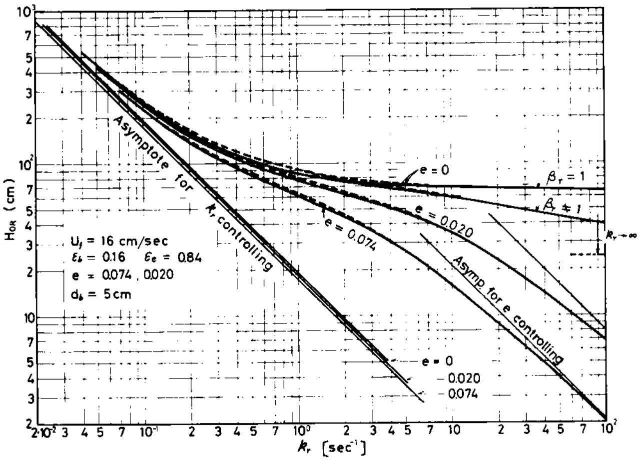

Fig. 4. Contributions of parameters on H_{oR}.

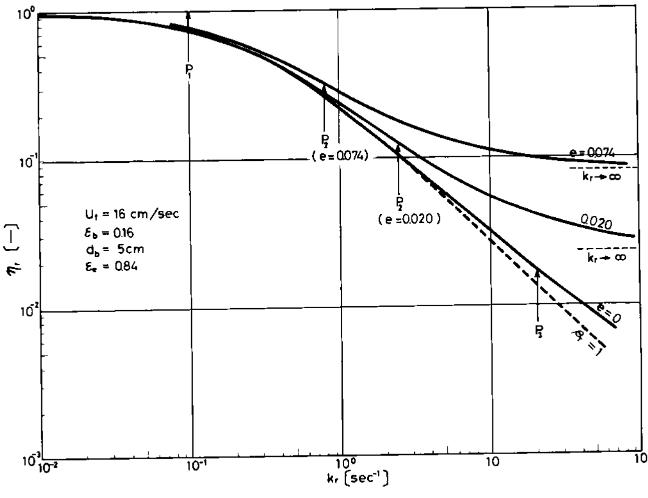

Fig. 5. Effect of the reaction rate on contact efficiency.

Table 2. Material and Enthalpy Balance Equations

Reaction	Equation (34)	Equation (35)	Equation (36)
$A \xrightarrow{1} B \xrightarrow{2} C$	$C_{eA} = \frac{N_{ob}C_{bA}}{N_{ob} + \phi_1\gamma_1}$ $C_{eB} = \frac{N_{ob}C_{bB} + \phi_1\gamma_1C_{eA}}{N_{ob} + \phi_2\gamma_2}$ $C_{eC} = \frac{\phi_2\gamma_2C_{eB} + N_{ob}C_{bC}}{N_{ob}}$	$\frac{dC_A}{dZ} - - \frac{Z_t - Z}{Z_t - 1}\phi_1\gamma_1^{1/\theta}C_A$ $\frac{dC_B}{dZ} = \frac{Z_t - Z}{Z_t - 1}(\phi_1\gamma_1^{1/\theta}C_A - \phi_2\gamma_2^{1/\theta}C_B)$ $\frac{dC_C}{dZ} = \frac{Z_t - Z}{Z_t - 1}\phi_2\gamma_2^{1/\theta}C_B$	$-\frac{d\theta}{dZ} - \Gamma(\theta - \theta_c) + \frac{Z_t - Z}{Z_t - 1}\Omega_1\phi_1\gamma_1^{1/\theta}C_A$ $+ \frac{Z_t - Z}{Z_t - 1}\Omega_2\phi_2\gamma_2^{1/\theta}C_B = 0$
$A \xrightarrow{1} X \xrightarrow{3} Y$ $\overset{2}{\searrow} P \quad \overset{4}{\searrow} Q$	$C_{eA} = \frac{N_{ob}C_{bA}}{N_{ob} + \phi_1\gamma_1 + \phi_2\gamma_2}$ $C_{eP} = \frac{N_{ob}C_{bP} + \phi_2\gamma_2C_{eA}}{N_{ob}}$ $C_{eX} = \frac{N_{ob}C_{bX} + \phi_1\gamma_1C_{eA}}{N_{ob} + \phi_3\gamma_3 + \phi_4\gamma_4}$ $C_{eQ} = \frac{N_{ob}C_{bQ} + \phi_4\gamma_4C_{eX}}{N_{ob}}$ $C_{eY} = \frac{N_{ob}C_{bY} + \phi_3\gamma_3C_{eY}}{N_{ob}}$	$-\frac{dC_A}{dZ} - \frac{Z_t - Z}{Z_t - 1}(\phi_1\gamma_1^{1/\theta} + \phi_2\gamma_2^{1/\theta})C_A = 0$ $-\frac{dC_P}{dZ} + \frac{Z_t - Z}{Z_t - 1}\phi_2\gamma_2^{1/\theta}C_A = 0$ $-\frac{dC_X}{dZ} + \frac{Z_t - Z}{Z_t - 1}(\phi_1\gamma_1^{1/\theta}C_A - (\phi_3\gamma_3^{1/\theta}$ $+ \phi_4\gamma_4^{1/\theta})C_X) = 0$ $-\frac{dC_Q}{dZ} + \frac{Z_t - Z}{Z_t - 1}\phi_4\gamma_4^{1/\theta}C_X = 0$ $-\frac{dC_Y}{dZ} + \frac{Z_t - Z}{Z_t - 1}\phi_3\gamma_3^{1/\theta}C_X = 0$	$-\frac{d\theta}{dZ} - \Gamma(\theta - \theta_c) - \frac{Z_t - Z}{Z_t - 1}\Omega\Big\{\phi_1\gamma_1^{1/\theta}C_A$ $+ \phi_2\gamma_2^{1/\theta}C_A + \phi_3\gamma_3^{1/\theta}C_X + \phi_4\gamma_4^{1/\theta}C_X\Big\} = 0$ ($\Omega = \Omega_1 = \ldots\ldots = \Omega_4$ is assumed.)
$A \overset{1}{\rightleftarrows} X$ $3 \overset{2}{\searrow} Y$	$C_{eA} = \frac{a_1C_{bA} + a_2b_1C_{bX}}{1 - a_2b_2}$ $C_{eX} = \frac{b_1C_{bX} + a_2b_2C_{bA}}{1 - a_2b_2}$ where, $a_1 = \frac{N_{ob}}{\phi_1\gamma_1 + \phi_3\gamma_3 + N_{ob}}, a_2 = \frac{\phi_1\gamma_1/K_R}{\phi_1\gamma_1 + \phi_3\gamma_3 + N_{ob}}$ $b_1 = \frac{N_{ob}}{\phi_1\gamma_1/K_R + N_{ob}}, b_2 = \frac{\phi_1\gamma_1}{\phi_1\gamma_1/K_R + N_{ob}}$	$-\frac{dC_A}{dZ} + \frac{Z_t - Z}{Z_t - 1}\Big\{-(\phi_1\gamma_1^{1/\theta} + \phi_3\gamma_3^{1/\theta})C_A$ $+ \frac{\phi_1\gamma_1^{1/\theta}}{K_R}\omega^{1-\frac{1}{\theta}}C_X\Big\} = 0$ $-\frac{dC_X}{dZ} + \frac{Z_t - Z}{Z_t - 1}\Big\{\phi_1\gamma_1^{1/\theta}\Big(C_A$ $-\frac{1}{K_R}\omega^{1-\frac{1}{\theta}}C_X\Big)\Big\} = 0$ $-\frac{dC_Y}{dZ} + \frac{Z_t - Z}{Z_t - 1}\phi_3\gamma_3^{1/\theta}C_A = 0$ ($\omega = \exp(-\Delta H_1/RT_f)$)	$-\frac{d\theta}{dZ} - \Gamma(\theta - \theta_c) + \Omega\Big\{-\frac{dC_A}{dZ}\Big\} = = 0$ ($\Omega = \Omega_1 = \Omega_3$ is assumed)

reactions, the dilute phase may cause the better selectivity due to better contact in the region. This effect was suggested by Furusaki (1973) and Miyauchi (1974b). In case of nonisothermal reactions there will be a different effect of the dilute phase. Mixing of gas and solids in the dilute phase is not sufficient, and this may cause a temperature distribution for exo or endothermic reactions in the phase. This temperature distribution may reveal a drastic effect on the selectivity of complex reactions. Discussion of this temperature effect is possibly important in the scale-up design of fluid bed reactors. To simplify our discussion, the following assumptions are made: that is, $\varphi \geqq 10$, $N_{rb} = 0$. Also it is assumed that the temperature in the dense phase is uniform. Then the material balance equation is simplified, for the dense phase,

$$\frac{dC_{bi}}{dZ} + N_{ob}(C_{bi} - C_{ei}) = 0 \tag{33}$$

$$N_{ob}(C_{bi} - C_{ei}) + \phi(C_{ei}) = 0 \tag{34}$$

and for the dilute phase

$$\frac{dC_i}{dZ} + \frac{Z - Z_t}{1 - Z_t}\phi(C_i) = 0 \tag{35}$$

where it is assumed that the catalyst density distribution varies linearly from $Z = 1$ to Z_t[*] (Furusaki, 1973). $\phi(C_i)$ is the term for the reaction and equals $\epsilon_e N_r C_i$ for the case of an isothermal first-order irreversible reaction and $(\epsilon_e k_r C_f^{n-1} L_f/U_f) \cdot C_i$ for nth order reactions. The enthalpy balance equation in the dilute phase is

$$-\frac{d\theta}{dZ} - \Gamma(\theta - \theta_c) + \frac{Z - Z_t}{1 - Z_t}\Omega\phi(C) = 0 \tag{36}$$

For usual fluid bed operations, the dilute phase is thermally insulated except when cold shot (Riley, 1959) is used to reduce the temperature rise in the dilute phase. In this case, the second term of Equation (36) can be neglected, and calculations are made for this case.

Case of Consecutive Reactions

The problem on the selectivity of B in consecutive reactions ($A \xrightarrow{1} B \xrightarrow{2} C$) is designated as the selectivity of Type III by Wheeler (1955). He examined the effect of mass transfer in the particles on the selectivity. In case of fluid bed reactors, the dense phase will be considered similarly as for the case of catalysis with mass transfer resistance. Hence, the selectivity of B will be greater for the case of larger N_{ob}. Moreover, in case of an isothermal reaction it will be greater if e is chosen larger because direct contact is assured in the dilute phase. For nonisothermal reactions the above expectation is not true because of the temperature distribution in the dilute phase.

For the case of the reactions in series ($A \xrightarrow{1} B \xrightarrow{2} C$), the rates of increase of A, B, C, are given by $R_A = -k_1C_A$, $R_B = k_1C_A - k_2C_B$, and $R_C = k_2C_B$, respectively. The k's are also expressed by the Arrhenius equation $k = A_r e^{-E/RT}$. Equations (34), (35), and (36) for this case are given in Table 2. Ω in Equation (36) is a nondimensional parameter relating to the heat of reaction, and the signature of Ω is +, 0, −, respectively, for exothermic, isothermal and endothermic reactions.

Calculations were made by a digital computer HITAC 8800/8700 of computation center of the University of Tokyo. Examples for isothermal reactions are given in Figure 6. In this case, the reaction rate of $B \rightarrow C$ is taken to be smaller than $A \rightarrow B$ by the factor of 10. For $N_{ob} < 3$, the yield of B is higher for $Z_t = 2$ than for $Z_t = 1$ (no catalysts in the dilute phase). This shows that the selectivity improves if one uses the efficiency of the dilute phase. Increasing the velocity of gas will enlarge the role of the dilute phase. Probably this is one way of increasing capacity.

Figure 7 shows a concentration profile for a nonisothermal reaction. Figure 8 shows the yield of B for $Z_t = 2$. For an exothermic reaction, and especially for $E_2 > 10$ kcal/mole, the yield of B decreases considerably. This is because the reaction $B \rightarrow C$ proceeds at higher temperatures in the dilute phase. Many oxidation reactions operated in fluid beds are exothermic reactions with $E_2 > E_1$ and $E_2 >> 10$ kcal/mole. In these reactions, it is not

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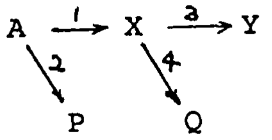
[*] This Z_t is not necessarily the same as one calculated from L_f of Figure 1 (compare Figure 13 of Furusaki, 1973).

profitable to enhance the role of the catalysts in the dilute phase so that U_f must be moderate or small in order to suppress the amount of catalyst in the dilute phase. Also, the use of internals, such as perforated plates, are helpful to depress the circulation in the dense phase and the carry over of catalysts into the dilute phase.

In case of endothermic reactions, as shown in the figure, the yields do not vary significantly from isothermal reactions. For $E_2 > 30$ kcal/mole, the use of the dilute phase becomes profitable. Since E_2 is usually large for the case of Type III, it may be desirable to use the dilute phase.

Denbigh's System with the Optimal Temperature Distribution

Denbigh (1958) proposed the following reaction system as the general case for organic reactions:



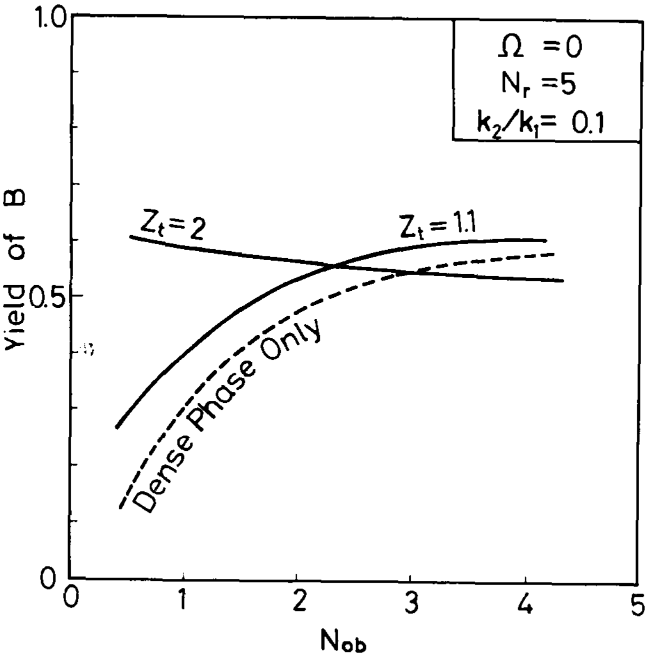


Fig. 6. Selectivity of isothermal reactions ($A \xrightarrow{1} B \xrightarrow{2} C$).

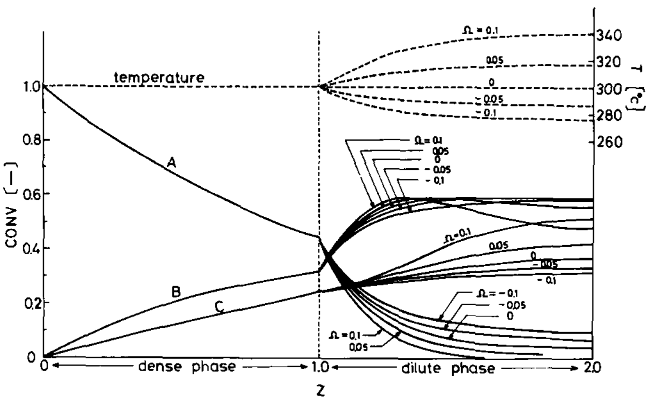


Fig. 7. Concentration profiles for consecutive reactions ($A \xrightarrow{1} B \xrightarrow{2} C$) $N_{ob} = 1$, $\phi_1 = 2 \times 10^8$, $E_1 = 20$ kcal/mole, $\phi_2 = 2 \times 10^7$, $E_2 = 20$ kcal/mole.

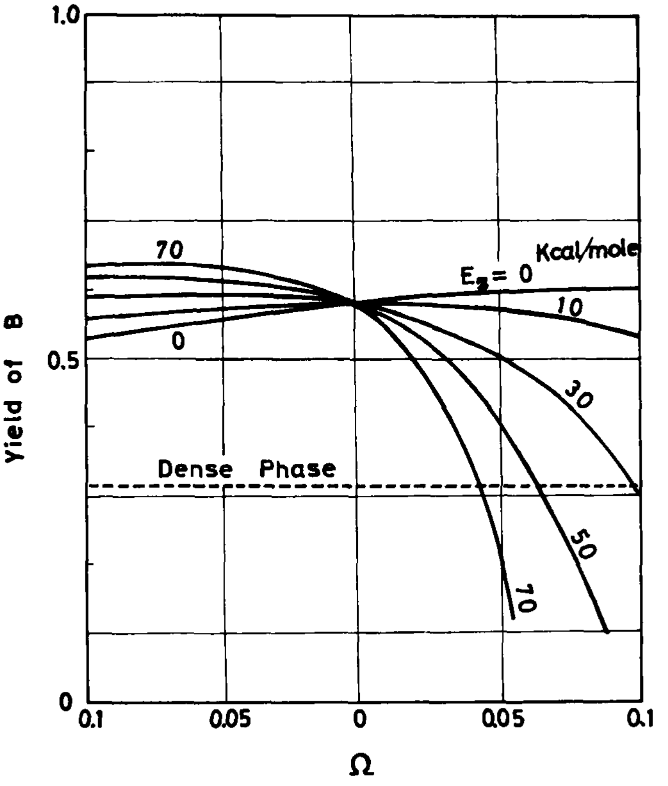


Fig. 8. Selectivity for non-isothermal consecutive reactions ($A \rightarrow B \rightarrow C$), $N_{ob} = 1$, $Z_t = 2$, $E_1 = 20$ kcal/mole, $\phi_1 = 2 \times 10^8$, $\phi_2 = \phi_1/10$.

This is a system with an optimal temperature distribution although the system in the previous section is one with an optimal residence time. In this system, the cases of (1) $E_1 < E_2$ and $E_3 > E_4$ or (2) $E_1 > E_2$ and $E_3 < E_4$ are especially interesting in considering the effect of the dilute phase. Reaction rates are expressed as $R_A = -(k_1 + k_2)C_A$, $R_P = k_2C_A$, $R_X = k_1C_A - (k_3 + k_4)C_X$, $R_Q = k_4C_X$, $R_Y = k_3C_X$ with Arrhenius relationships. Material balance equations are shown in Table 2.

For the case of $E_1 < E_2$, $E_3 > E_4$, Denbigh showed a considerable increase of the yield of Y in continuous stirred tank reactors (CSTR) by maintaining the temperature low in the first vessel and higher in the succeeding vessels. In fluid beds, one cannot control the temperature distribution, but the temperature changes between 0 ~ 30°C in the dilute phase. Hence the effect of temperature distribution is not as large in fluid beds as in a CSTR, although it is found to be preferable for exothermic reactions ($E_1 < E_2$, $E_3 > E_4$). As for endothermic reactions, the opposite situation is found. However, the reaction cools down in the dilute phase so that the yield does not decrease significantly.

Calculations for $E_1 = E_3 = 30$ kcal/mole, $E_2 = 50$, $E_4 = 10$ kcal/mole show a considerable increase in yield of Y for the case of exothermic reactions such as $\Omega = 0.1$. An example is shown in Figures 9 and 10. Values of ϕ are determined so that at $\theta = 1$, $\epsilon_e N_{r1} = \epsilon_e N_{r3} = 5$ and $\epsilon_e N_{r2} = \epsilon_e N_{r4} = 0.5$. The difference between solid lines and broken lines shows the extent of reaction in the dilute phase. The good effect of the dilute phase is obvious for $\Omega = 0.1$. For endothermic reactions ($\Omega = -0.1$), the effect is small. But even if the contact in the dense phase is improved (large N_{ob}), calculations show that the yield does not improve. For example, the yield is 72.6% for $N_{ob} = 1$ and 73.7% for $N_{ob} = 10$. So, there is no large difference between the suppression and enhancement of the dilute phase effect.

For the case of endothermic reactions, $E_1 > E_2$, $E_3 < E_4$, it is preferable to use the dilute phase. But since the reaction quenches itself in this phase, the selectivity does not improve significantly. In case of exothermic reactions, a high conversion in the dense phase is preferable. But calculations show that improvement by increasing N_{ob} is not significant because the yield of P increases although that of Q decreases.

Generally speaking for the Denbigh system with optimal temperature distribution, reactions in the dilute phase do not make the selectivity unfavorable. Therefore, operation with a large U_f may possibly be the economically optimal step. If Y reacts further into byproducts, the same discussion can be applied as in consecutive reactions.

Parallel Reactions with an Equilibrium

The parallel reaction $A \genfrac{}{}{0pt}{}{\nearrow X}{\searrow Y}$ is another important reaction system. If the orders of the two reactions are the same, the selectivity does not depend upon the type of reactor. An example of an industrially important parallel reactions is $A \genfrac{}{}{0pt}{}{\overset{1}{\underset{2}{\rightleftarrows}} X}{\underset{3}{\searrow} Y}$ (X is the required product (Denbigh, 1965).

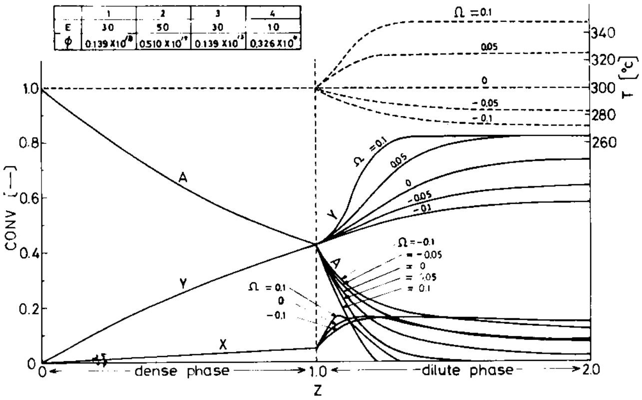


Fig. 9. Concentration profiles for the Denbigh'a complex reaction systems, $A + B \xrightarrow{1} X \xrightarrow{3} Y$ (Y is the required product), $N_{ob} = 1$. $B \xrightarrow{2} P$, $X \xrightarrow{4} Q$

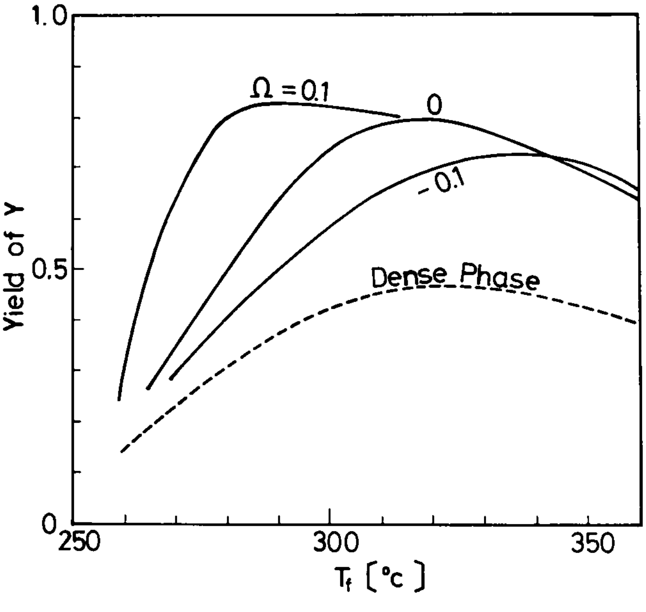


Fig. 10. Selectivity for the Denbigh's system, $E_1 = E_3 = 30$ kcal/mole, $E_2 = 50$ kcal/mole, $E_4 = 10$ kcal/mole, $Z_t = 2$.

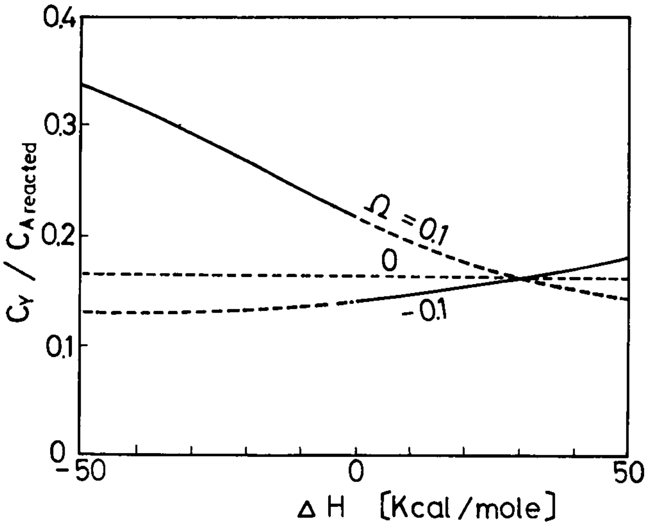


Fig. 11. Effect of the heat of reaction on the selectivity for the reaction with an equilibrium, $A \underset{2}{\overset{1}{\rightleftarrows}} X$ (X is the required product), $A \xrightarrow{3} Y$, $N_{ob} = 1$, $\phi_1 = 1.387 \times 10^{12}$, $\phi_3 = \phi_1/10$, $E_1 = E_3 = 30$ kcal/mole, $K_R = 10$, $Z_t = 3$.

Examples of this reaction system are the hydrocarbon and methanol synthesis reactions. Packed bed reactors with an internal cooling system are used for this type of reaction. X produced by the reaction should be removed from the reactor as soon as it is produced. Fluid beds with large N_{ob} are preferable for this case.

Material and enthalpy balances for $R_A = -(k_1 + k_3)C_A + k_2C_X$, $R_X = k_1C_A - k_2C_X$, and $R_Y = k_3C_A$ are given in Table 2. The temperature effect of this example is examined in Figure 11. If the reaction 1 is exothermic ($\Delta H_1 < 0$), equilibrium is unfavorable for overall exothermic reaction systems ($\Omega = 0.1$). If $\Delta H_1 > 0$, the tendency is opposite although the effect is not significant. This is because the decrease of temperature in the dilute phase quenches the progress of reaction into Y. Figure 12 shows the effect of N_{ob} on the yield of Y (undesired product). It shows that in this reaction system it is favorable to make N_{ob} large in order to raise the conversion in the dense phase.

DISCUSSION

In fluid bed reactors, the effect of catalyst particles dispersed in the dilute phase cannot be neglected. The history of the progress of fluid beds is interesting from this point of view. One example of development is fluidized catalytic cracking (FCC). In this case, reactors have become slender and the superficial velocity of gas has become larger. It is almost like a transport bed since most of the reaction occurs in the riser (Yoshida, 1973).

The other direction of development is partial oxidation reactions like ammoxidation or oxichlorination. In these cases, the velocities of the gas are moderate and reactors have internals like perforated plates in order to avoid circulation in the dense phase. Organic reactions have the property of consecutive reaction since products react further into byproducts. In case of FCC, product naphtha will be cracked further into carbon; in case of partial oxidation products will be oxidized further into CO_2 and H_2O. An analysis by the simplest model ($A \rightarrow B \rightarrow C$) shows that for the endothermic or isothermal reactions, one may use the dilute phase but for the exothermic reactions one must avoid reaction occurring in the dilute phase. Of course,

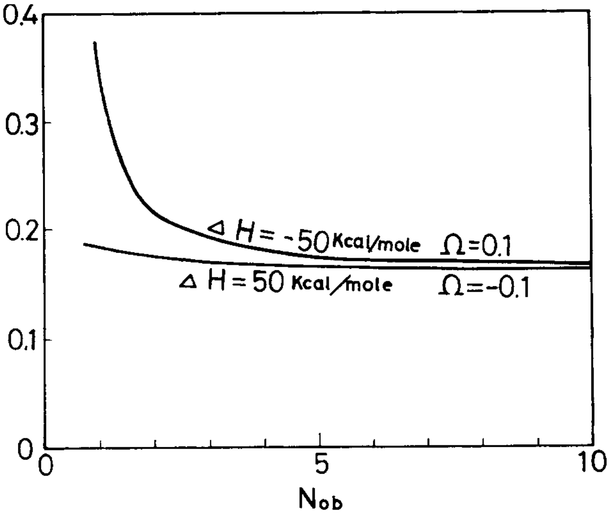


Fig. 12. Effect of N_{ob} on the yield of the byproduct $Y(A \overset{1}{\rightleftarrows} X), \searrow^{2}_{3} Y$ $\phi_1 = 1.387 \times 10^{12}$, $\phi_3 = \phi_1/10$, $E_1 = E_3 = 30$ kcal/mole, $Z_t = 3$, $K_R = 10$.

for simple exothermic reactions without byproduct, the dilute phase will give larger contact efficiency and a higher yield of products.

Calculations show when the use of the dilute phase is profitable, the decision depends upon the reaction. This point may be important in deciding policies of scale-up of fluid bed reactors.

From an industrial viewpoint, the control of temperature and amount of catalyst in the dilute phase is important. This is done by the appropriate choice of gas velocity, catalyst activity, catalyst size and density, and also appropriate design of reactor internals. The amount of catalyst particles in the dilute phase is reported by Miyauchi (1974a) and Tsutsui (1974). However, the study on the dilute phase in fluid beds is rather limited and more studies will be necessary on the dilute phase, including the transition zone (Figure 1).

NOTATION

A = area for heat transfer between the dilute phase and wall, cm^2/cm^3
A_r = frequency factor for the Arrhenius equation, s^{-1}
a_b = mass transfer area between bubble and emulsion, cm^2/cm^3
a_{ex} = area for the exchange of material between the ascending and descending emulsion, cm^2/cm^3
C = concentration, g mole/cm^3
$C_f{}^0$ = concentration of the inlet gas, g mole/cm^3
C_{f1} = concentration of gas at the top of the dense phase, g mole/cm^3
C_{fo} = concentration of the outlet gas, g mole/cm^3
c_p = heat capacity of the gas, cal/g · C
D_{eff} = effective diffusivity in the emulsion or cloud, $\epsilon_{fe} D_G/\chi$, cm^2/s
D_G = molecular diffusivity, cm^2/s
d_b = diameter of bubbles, cm
d_{vs} = surface mean diameter of bubbles, cm
E = activation energy, kcal/g mole
e = averaged amount of catalyst particles in the dilute phase, $\int_1^{Z_t} \epsilon_e dZ$, dimensionless
g = gravitational acceleration, cm/s^2
ΔH = heat of reaction, kcal/g mole
H_b = height of a transfer unit for the bubble phase, $U_f/k_b a_b$, cm
H_e = height of a transfer unit for the emulsion phase, $U_f/k_e a_b$, cm
$H_{ob})_{\text{app}}$ = apparent overall height of a transfer unit between bubble and emulsion, cm
H_{ob} = overall height of a transfer unit for mass transfer, $U_f/k_{ob} a_b$, cm
H_{or} = overall height of a transfer unit for the dense phase, U_f/k_{or}, cm
H_{oR} = overall height of a transfer unit for the whole bed, U_f/k_{oR}, cm
H_r = height of a reaction unit, U_f/k_r, cm
J = correction parameter for the cloud, refer to Equation (5), dimensionless
K = reaction rate constant for the fixed bed, s^{-1}
K_R = equilibrium constant, dimensionless
k_b = mass transfer coefficient in the bubble phase, cm/s
k_e = mass transfer coefficient in the emulsion phase, cm/s
k_{oR} = $k_{or} + (\mu\epsilon_b + e) k_r$, s^{-1}
k_{or} = $\dfrac{1}{\dfrac{1}{k_{ob} a_b} + \dfrac{1}{\xi \epsilon_e k_r}}$, s^{-1}
k_{ob} = overall mass transfer coefficient between the bubble and emulsion phase, cm/s
k_r = reaction rate constant, s^{-1}
L = height, cm
L_f = height of the dense phase, cm
L_q = height of the quiescent bed, cm
L_t = total bed height, cm
m = adsorption equilibrium constant for the emulsion, $\epsilon_{fe} + m_s (1 - \epsilon_{fe})$, dimensionless
m_H = $\sqrt{k_r D_{\text{eff}}}/k_e$, dimensionless
m_s = adsoprtion equilibrium constant for catalyst particles, dimensionless
N_{ex} = $m k_{ex} a_{ex} L_f / U_f$, dimensionless
N_{ob} = number of transfer units between bubble and emulsion, $k_{ob} a_b L_f / U_f$, dimensionless
$(N_{ob})_{\text{app}}$ refer to Equation (30), dimensionless
N_{oR} = overall number of a transfer unit, $N_{or} + (\mu\epsilon_b + e) N_r$, dimensionless
N_{or} refer to Equation (11), dimensionless
$(N_{oR})_{\text{FIX}} = N_{oR}$ for the fixed bed, $K L_q / U_f$, dimensionless
N_r = $k_r L_f / U_f$, dimensionless
P_e = $m \delta^2 / 4 r_b D_{\text{eff}}$, dimensionless
R_A, R_B . . . = rate of the reaction to produce A, B, . . ., g mole/cm^3 s
T = temperature, °C, °K
T_c = temperature of the coolant, °C
T_f = temperature in the dense phase, °C
U = overall heat transfer coefficient, cal/cm^2 °C s
U_e = superficial circulation velocity of the emulsion phase, cm/s
U_f = superficial velocity of gas, cm/s
u_b = rising velocity of bubbles relative to the emulsion, cm/s
U_{mf} = superficial velocity of gas at the incipient fluidization, cm/s
Z = height of the bed, L/L_f, dimensionless
Z_t = total bed height, L_t/L_f, dimensionless
k_{ex} = mass exchange coefficient between the ascending and descending emulsion, cm/s

Greek Letters

α = $\epsilon_{mf} u_b / u_{mf}$, dimensionless
β = Hatta number, dimensionless
β_r = modified Hatta number for the emulsion, dimensionless
Γ = $U A L_f / U_f c_p \rho_f$, dimensionless

γ = $\exp(-E/RT_f)$, dimensionless

δ = thickness of the cloud,

$$\frac{d_b}{2}\left\{\left(\frac{\alpha}{\alpha-1}\right)^{1/3}-1\right\}\approx\frac{d_b}{6\alpha}, \text{cm}$$

ϵ_b = volume fraction of the bubble phase, dimensionless

ϵ_e = volume fraction of the emulsion phase, dimensionless

ϵ_e^* = $\epsilon_e + \mu\epsilon_b + e$, dimensionless

ϵ_{fe} = void fraction of the emulsion phase, dimensionless

ϵ_{mf} = void fraction at the incipient fluidization, dimensionless

η_r = contact efficiency, dimensionless

θ = T/T_f, dimensionless

θ_c = T_c/T_f, dimensionless

μ = fractional amount of catalyst particles in the bubble phase, dimensionless

ξ = effective fraction of catalyst, refer to Equation (12), dimensionless

Ω = $\dfrac{(-\Delta H)C_f^0}{c_p\rho_f T_f}$, dimensionless

ρ_f = density of gas, g/cm^3

τ_b = local surface renewal time between bubble and emulsion, s

ϕ = $\epsilon_e L_f A_r/U_f$, dimensionless

$\phi(c)$ = nondimensional reaction rate

χ = tortuosity factor, dimensionless

φ refer to Equation (7), dimensionless

Subscripts

$1, 2, \ldots$ = reactions

$A, B, \ldots$ = substances

b = bubble phase

e = emulsion phase

f = gas

s = solid

eu, ed = ascending and descending emulsion, respectively

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