

# INCLUSIVE TWO-PHASE MODEL FOR FLUIDIZED-BED REACTOR

IWA0 MUCHI

*Department of Iron and Steel Engineering*

(Received May 31, 1965)

## Introduction

In order to evaluate the possible conversion of catalytic reaction in gaseous fluidized-bed reactor, various two-phase models were derived by many investigators. Before making use of two-phase model, it is preferable to understand sufficiently the characteristics of this model.

A generalized mathematical model which include various two-phase models presented hitherto is derived here from the concept based on the two-phase theory. By analyzing inclusive two-phase model, the relations with other models and the effects of operating conditions on the conversion of fluidized-bed reactor were investigated, and thus, the characteristics of two-phase model were clarified in this report.

### 1. Inclusive Two-phase Model

It is imagined that aggregative fluidized-bed reactor consists of an emulsion phase and a dilute phase. And these phases construct a parallel reactor with interchange of gas in axial direction as illustrated in Fig. 1.

In deriving the inclusive two-phase model, it has been assumed: that the radial distributions of the concentration of reactant gas in each phase are uniform; the behaviours of gas flow is piston flow in dilute phase and incomplete mixing in emulsion phase; there is a cross flow of gases through the interface between both phases, and the volume rate of the cross flow of gas and the effective area of interface are respectively constant in every height from a bottom of the bed; catalyst particles are contained in both phases and the irreversible reactions which take place under isothermal conditions in

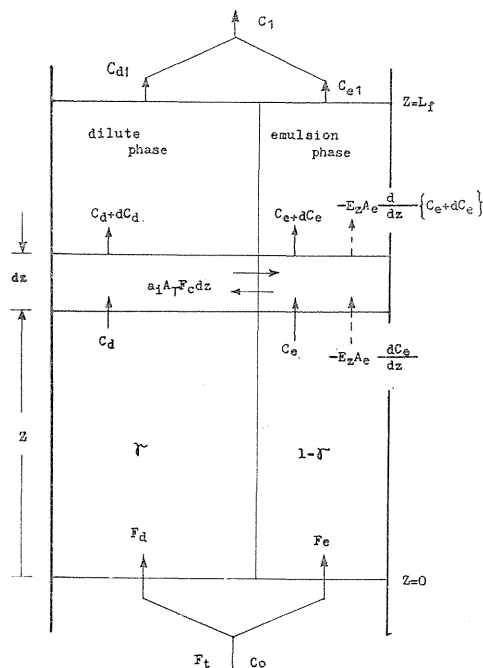


FIG. 1. Two-phase model of fluidized bed

and the irreversible reactions which take place under isothermal conditions in

each phase are first order with respect to the gaseous reactant; and longitudinal distributions of bed density are constant in each phase.

Under these assumptions above-mentioned, the material balances around the differential elements of the bed in each phase can then be written as Equation (1) for dilute phase and Equation (2) for emulsion phase.

$$\frac{dC_d}{dz} + \frac{a_i F_c A_T}{F_d} (C_d - C_e) + \frac{\gamma k W_s}{F_d L_f} C_d = 0 \quad (1)$$

$$\frac{d^2 C_e}{dz^2} - \frac{F_e}{A_e E_z} \frac{dC_e}{dz} + \frac{a_i F_c A_T}{A_e E_z} (C_d - C_e) - \frac{(1-\gamma) k W_s}{A_e E_z L_f} C_e = 0 \quad (2)$$

The boundary conditions are assumed to be :

$$C_d = C_0 \quad \text{at} \quad z = 0 + \quad (3)$$

$$dC_e/dz = - (F_e/A_e E_z) (C_0 - C_e^0) \quad \text{at} \quad z = 0 + \quad (4)$$

$$dC_e/dz = 0 \quad \text{at} \quad z = L_f \quad (5)$$

Solving the fundamental equations (1) and (2) with these boundary conditions shown here, concentrations of reactant at the top of dilute and emulsion phases are obtained as follows :

$$C_{d1}/C_0 = \sum_{j=1}^3 \{ b A_j / (a + \alpha_j) \} (e^{\alpha_j} - e^{-a}) + e^{-a} \quad (6)$$

$$C_{e1}/C_0 = \sum_{j=1}^3 A_j e^{\alpha_j} \quad (7)$$

where  $\alpha_1 \sim \alpha_3$  are the roots of Equation (9).

The exit concentration of reactant is given by :

$$C_1 = (C_{d1} F_d + C_{e1} F_e) / F_t \quad (8)$$

$$y^3 + (a - m)y^2 - m(a + g)y - m(ag - bf) = 0 \quad (9)$$

$A_1$ ,  $A_2$ , and  $A_3$  in Equations (6) and (7) are denoted as follows :

$$A_1 = (m/\lambda) [ \{ \alpha_1 \alpha_2 + (a+f)(m-\alpha_3) \} \alpha_2 e^{\alpha_2} - \{ \alpha_3 \alpha_1 + (a+f)(m-\alpha_2) \} \alpha_3 e^{\alpha_3} ] \quad (10)$$

$$A_2 = (m/\lambda) [ \{ \alpha_2 \alpha_3 + (a+f)(m-\alpha_1) \} \alpha_2 e^{\alpha_3} - \{ \alpha_1 \alpha_2 + (a+f)(m-\alpha_3) \} \alpha_1 e^{\alpha_1} ] \quad (11)$$

$$A_3 = (m/\lambda) [ \{ \alpha_3 \alpha_1 + (a+f)(m-\alpha_2) \} \alpha_1 e^{\alpha_1} - \{ \alpha_2 \alpha_3 + (a+f)(m-\alpha_1) \} \alpha_2 e^{\alpha_2} ] \quad (12)$$

where  $\lambda$  is defined as

$$\lambda = (\alpha_3 - \alpha_2)(a + \alpha_1)\alpha_1^2 \cdot e^{\alpha_1} + (\alpha_1 - \alpha_3)(a + \alpha_2)\alpha_2^2 \cdot e^{\alpha_2} + (\alpha_2 - \alpha_1)(a + \alpha_3)\alpha_3^2 \cdot e^{\alpha_3} \quad (13)$$

and  $a$ ,  $b$ ,  $f$ ,  $g$ , and  $m$  are denoted as follows :

$$\begin{aligned} a &= (F_{cr} + \gamma X) / F_{dr}, & b &= F_{cr} / F_{dr}, & f &= F_{cr} / F_{er}, & g &= \{ F_{cr} + (1-\gamma) X \} / F_{er}, \\ m &= F_e L_f / A_e E_z = u_e L_f / E_z = 2 U_e, & X &= k W_s / F_t, & F_{cr} &= a_i F_c A_T L_f / F_t, \\ F_{dr} &= F_d / F_t, & F_{er} &= F_e / F_t \end{aligned}$$

Then, fraction conversion can be obtained as follows :

$$\begin{aligned}\eta &= 1 - (C_1/C_0) \\ &= 1 - \frac{b}{1+(b/f)} \left\{ \sum_{j=1}^3 \left( \frac{1}{a+\alpha_j} + \frac{1}{f} \right) A_j e^{\alpha_j} + \left( \frac{1}{b} - \sum_{j=1}^3 \frac{1}{a+\alpha_j} A_j \right) e^{-a} \right\}\end{aligned}\quad (14)$$

Equation (14) is the inclusive two-phase model and various two-phase models presented hitherto can be included in this equation shown above.

Attempt to derive mathematically a general two-phase model from the unified standpoint of view was investigated by Kobayashi and Arai<sup>1)</sup>, but the perfect solution was not shown.

May<sup>2)</sup> presented a mathematical model which was based on the assumptions that there was no catalyst particle in dilute phase, gas in dilute phase flowed through in piston flow, and in emulsion phase the backmixing of gas took place.

Results obtained from putting  $\tau=0$  in Equation (14) of the inclusive two-phase model agree perfectly with the mathematical model of May<sup>2)</sup>.

## 2. Simplification of the Inclusive Two-phase Model

Making  $E_z \rightarrow 0$  in Equation (14) of the inclusive two-phase model, then Equation (15) of P·P-type two-phase model, in which gases in both phases flow through in piston flow, can be obtained.

$$\eta = 1 - \frac{1}{1+(b/f)} \left( \frac{b/f}{\beta_1 - \beta_2} \right) \left\{ \frac{(a+f+\beta_1)^2}{a+\beta_1} e^{\beta_1} - \frac{(a+f+\beta_2)^2}{a+\beta_2} e^{\beta_2} \right\}\quad (15)$$

where  $\beta_1$  and  $\beta_2$  are the roots of Equation (16).

$$y^2 + (a+g)y + (ag-bf) = 0\quad (16)$$

Substituting the solution of the fundamental equations (1) and (17) with boundary conditions: Equations (18) and (19), into Equation (8), then Equation (15) can also be obtained.

$$\frac{dC_e}{dz} + \frac{a_i F_r A_r}{F_e} (C_e - C_d) + \frac{(1-\tau)kW_s}{F_e L_f} C_e = 0\quad (17)$$

$$C_d = C_e = C_0 \quad \text{at } z = 0\quad (18)$$

$$C_d = C_{d1}, \quad C_e = C_{e1} \quad \text{at } z = L_f\quad (19)$$

Letting  $E_z \rightarrow \infty$  in Equation (14), then Equation (20) of P·M-type two-phase model, in which gas in emulsion phase is in complete mixing, can be obtained.

$$\eta = 1 - \frac{1}{1+(b/f)} \left[ e^{-a} + \frac{(b/f) \{ f(1-e^{-a}) + a \}^2}{a^2(1+g) + bf(1-a-e^{-a})} \right]\quad (20)$$

Equation (20) can also be derived from the results of combining Equation (8) with the solution of the fundamental equations (1) and (21), with boundary conditions: Equations (22) ~ (24).

$$F_e(C_{e1} - C_0) + a_i F_c A_T \int_0^{L_f} (C_{e1} - C_d) dz + (1 - \gamma) k W_s C_{e1} = 0 \quad (21)$$

$$C_d = C_0 \quad \text{at} \quad z = 0 \quad (22)$$

$$C_d = C_{d1} \quad \text{at} \quad z = L_f \quad (23)$$

$$C_e = C_{e1} \quad \text{for} \quad z = 0 + \sim L_f \quad (24)$$

As an extreme case, it may be favourable to imagine a parallel flow-reactor model having no gas-interchange between both phases.

Making  $F_{cr} \rightarrow 0$  in Equation (14), then Equation (25) of parallel flow-reactor model can be obtained.

$$\eta = 1 - F_{dr} \cdot \exp\left(-\frac{\gamma X}{F_{dr}}\right) - \frac{4 P F_{er}}{Q^2 \cdot \exp(-U_e R) - R^2 \cdot \exp(-U_e Q)} \quad (25)$$

where  $P \equiv \sqrt{1 + 2(1 - \gamma)X / F_{er} U_e}$ ,  $U_e \equiv u_e L_f / 2 E_z$ ,  $Q \equiv 1 + P$ ,  $R \equiv 1 - P$

This model represented by Equation (25) consists of two independent reactors. Dilute-phase reactor is piston-flow type and emulsion-phase reactor is incomplete-mixing type. The second term in the right-hand side of Equation (25) represents the conversion in dilute phase and the third term represents the conversion in emulsion phase.

Letting  $F_{cr} \rightarrow 0$  in Equation (15) of P·P-type two-phase model, then Equation (26) of P·P-type parallel-flow reactor model can be obtained.

$$\eta = 1 - F_{dr} \cdot \exp\left(-\frac{\gamma X}{F_{dr}}\right) - F_{er} \cdot \exp\left\{-\frac{(1 - \gamma)X}{F_{er}}\right\} \quad (26)$$

Equation (26) can also be obtained by substituting  $E_z \rightarrow 0$  in Eq. (25).

Making  $F_{cr} \rightarrow 0$  in Equation (20) of P·M-type two-phase model, then Equation (27) of P·M-type parallel-flow reactor model can be obtained.

$$\eta = 1 - F_{dr} \cdot \exp\left(-\frac{\gamma X}{F_{dr}}\right) - F_{er} \left\{1 + \frac{(1 - \gamma)X}{F_{er}}\right\} \quad (27)$$

Equation (27) can also be obtained by substituting  $E_z \rightarrow \infty$  in Equation (25).

Putting  $\gamma = 0$ ,  $F_{er} = 1$ ,  $u_e = u$ , and making  $F_{dr} \rightarrow 0$  in Equation (14) of inclusive two-phase model, then Equation (28) of a homogeneous phase model derived by Danckwerts<sup>3)</sup> and Yagi and Miyauchi<sup>4)</sup> can be obtained.

$$\eta = 1 - 4 P_h / \{Q_h^2 \cdot \exp(-UR_h) - R_h^2 \cdot \exp(-UQ_h)\} \quad (28)$$

where  $P_h \equiv \sqrt{1 + 2X/U}$ ,  $U \equiv u L_f / 2 E_z$ ,  $Q_h \equiv 1 + P_h$ ,  $R_h \equiv 1 - P_h$

And, Equation (28) can be led by substitution the same values of  $\gamma$ ,  $F_{er}$ ,  $u_e$ ,  $F_{dr}$  as shown above in Equation (25).

Putting  $\gamma = 0.5$ ,  $F_e = F_d$  in Equation (15), then Equation (29) which represents the homogeneous phase model of piston-flow type can be derived.

$$\eta = 1 - \exp(-X) \quad (29)$$

Putting  $\gamma = 0$ ,  $F_d = 0$  in Equation (20), then Equation (30) which represents the homogeneous phase model of complete-mixing type can be derived.

$$\eta = 1 - 1/(1 + X) \quad (30)$$

When  $\gamma \neq 0$ ,  $\eta \rightarrow 1$  as  $X \rightarrow \infty$  in Equation (14), but when  $\gamma = 0$ , letting  $X \rightarrow \infty$ , then Equation (31) yields.

$$\eta = 1 - F_{dr} \cdot \exp(-F_{cr}/F_{dr}) \quad (31)$$

These analyses imply that, when there are catalyst particles in dilute phase, conversion tend to 100% as increase in  $X$ , but when there is no catalyst in the phase, conversion tend to a lower value than 100%.

### 3. Evaluation of Conversion

Assuming that emulsion phase remain under conditions similar to those presented at incipient fluidization, then  $A_e$ ,  $F_{er}$ ,  $F_{dr}$ ,  $m$  can be represented, as follows:

$$A_e = \varepsilon_{mf}(1 - \gamma)(L_{mf}/L_f)A_r \quad (32)$$

$$F_{er} = F_e/F_t = (1 - \gamma)(u_{mf}L_{mf}/u_0L_f) \quad (33)$$

$$F_{dr} = F_d/F_t = 1 - F_{er} \quad (34)$$

$$m = u_e L_f / E_z = u_{mf} L_f / \varepsilon_{mf} E_z \quad (35)$$

Provided that the principal unknown factors such as  $L_f$ ,  $k$ ,  $a_i F_c$  (or  $F_{cr}$ ),  $\gamma$ ,  $E_z$  can be previously determined, the conversion would be estimated from Equation (14). In particular, it is very difficult to determine the following factors:  $a_i F_c$ ,  $\gamma$ ,  $E_z$ . But, in order to predict a practical conversion by means of inclusive two-phase model, it is necessary to present these unknown factors as the functions of operating conditions.

Observed values of  $\gamma$  by Towe, *et al.*<sup>5)</sup> were smaller than about 0.006, if so, it might be guessed from the theoretical ground that the magnitude of  $\gamma$  would not affect the degree of conversion in catalytic reaction.

In order to clarify the effect of back-mixing in emulsion phase on the overall conversion, the variations of  $Y \equiv (\eta_{P,P} - \eta_{P,M}) / \eta_{P,P}$  with  $X$  are illustrated in Fig. 2 and 3. From these graphs, it is seen that the curve has a maximum value at a certain value of  $X$ , and the increase in the height of curve implies that the effect of the magnitude of  $E_z$  on conversion becomes remarkable. When  $X < 0.05$  or  $X > 10$ , it is found that the difference between conversions resulted from Equation (15) and those from Equation (20) might become lower than several per-centage.

The relations of  $X$  to conversions of various two-phase models, homogeneous models, and modified two-phase model given by Davidson and Harrison<sup>6)</sup> are shown in Fig. 4, in terms of parameters of  $F_{er}$ ,  $F_{cr}$ , and  $\gamma$ . Curves for the case of piston flow and complete mixing are included for comparison.

When  $X=1$ , the effect of  $F_{cr}$  on  $\eta$  is illustrated in Fig. 5, in terms of parameter of  $\gamma$ . The increase in  $\eta$  as increase in  $F_{cr}$  becomes remarkable at low  $F_{cr}$  level than at high. It is guessed that the effect of  $\gamma$  may be diminished as increase in  $F_{cr}$ , and the curves tend to the value of  $\eta$  indicated by arrows.

Adopting the experimental data obtained by Massimilla *et al.*,<sup>7)</sup> Shen *et al.*,<sup>8)</sup> Johnstone *et al.*,<sup>9)</sup> and Mathis *et al.*,<sup>10)</sup> the calculated results of  $F_{cr}$  from inclusive two-phase model are given in Table 1. It may be considered that back-mixing

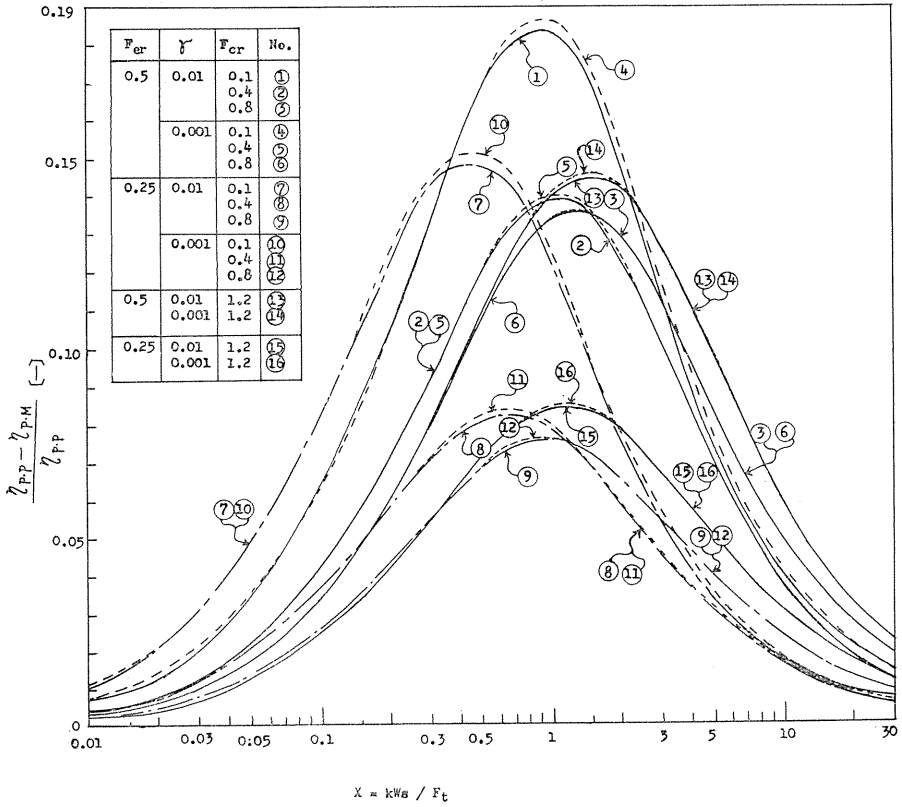


FIG. 2. Effect of  $E_z$  on conversion for two-phase model

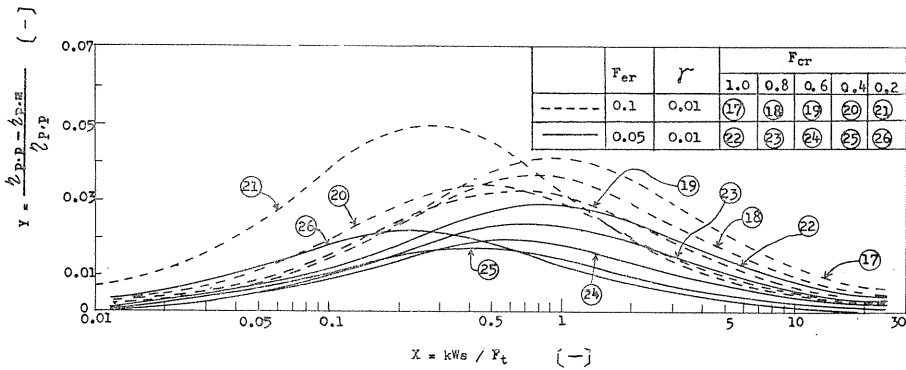


FIG. 3. Effect of  $E_z$  on conversion for two-phase model

in the experiments performed by Massimilla *et al.* and by Mathis *et al.* had negligible effects, because the values of  $X$  in the former were very small and those in the latter were very large.

Whereas, basing on the levels of  $X$  given by Shen *et al.*, it might be guessed that some effects of  $E_z$  would appear. Considering that the values of  $F_{cr}$  of Shen

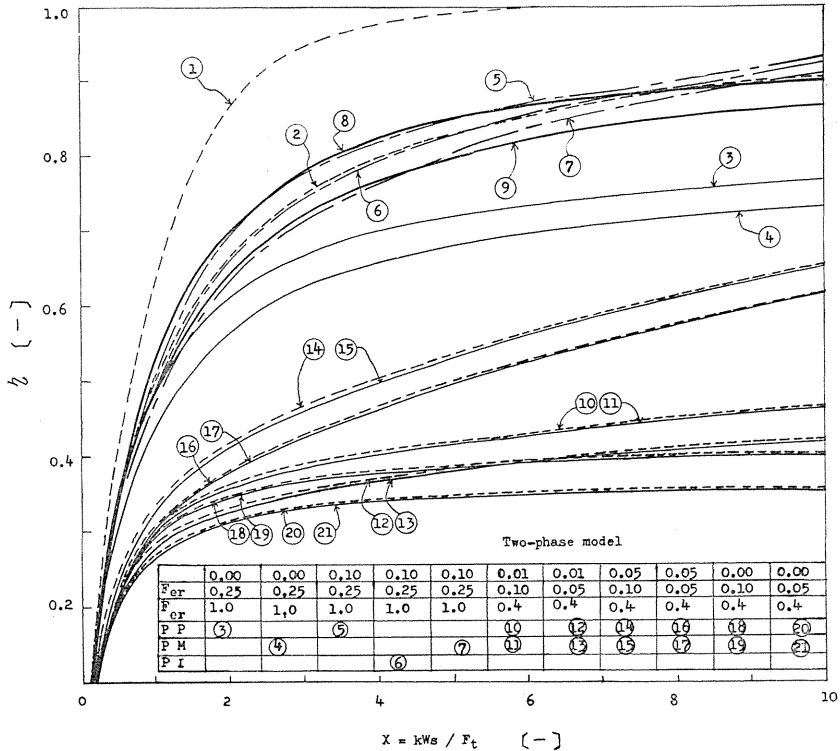
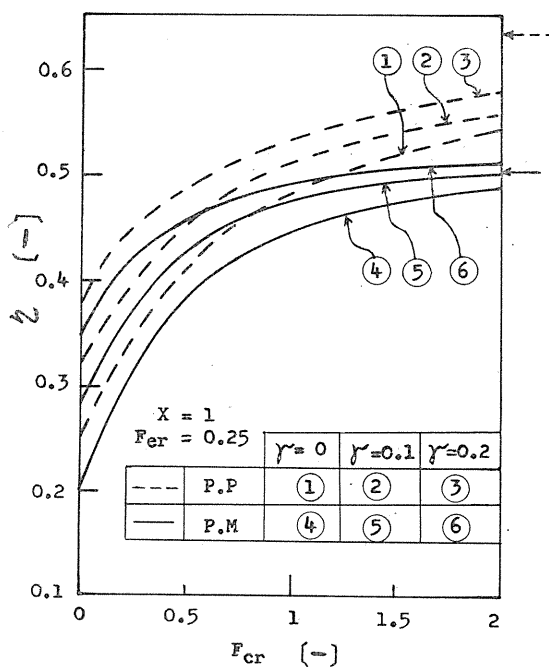


FIG. 4. Homogeneous-phase model:

- ① piston flow, ② complete mixing
- Davidson and Harrison's model:
- ③ piston flow in emulsion phase
- ④ complete mixing in emulsion phase

TABLE 1. Estimation of cross-flow rate of gas  
 Run No. 1-4: Massimilla *et al.*<sup>7)</sup>; No. 5-12: Mathis *et al.*<sup>10)</sup>; No. 13-15: Shen *et al.*<sup>8)9)</sup>

Run No.	$\eta$	$u_0$ (cm/sec)	$L_f$ (cm)	$F_{er}$	$X$	$F_{er}$
1	0.48	8.43	26.5	0.154	0.087	0.60
2	0.42	11.8	29.6	0.098	0.062	0.50
3	0.35	16.4	32.6	0.094	0.045	0.43
4	0.30	20.4	36.8	0.046	0.036	0.40
5	0.59	3.66	8.07	0.119	22.61	0.48
6	0.55	6.40	8.07	0.058	10.95	0.56
7	0.48	3.66	10.53	0.119	29.36	0.40
8	0.46	6.10	10.53	0.062	15.39	0.53
9	0.48	3.66	5.29	0.119	14.82	0.40
10	0.51	6.71	5.29	0.053	6.66	0.605
11	0.64	3.66	15.88	0.119	44.35	0.71
12	0.65	6.10	15.9	0.062	23.29	0.90
13	0.26	1.27	29.7	0.346	0.341	0.10
14	0.14	2.46	32.6	0.152	0.176	0.05
15	0.10	3.41	34.2	0.104	0.127	0.05

FIG. 5.  $\eta$  vs.  $F_{cr}$  at  $X=1$ 

*et al.* are very small in comparison with the others, conversions were calculated by means of parallel-flow model. The results indicate that the conversions were closer to those expected for P·P-type model than for P·M-type, as shown in Table 2. It means that the values  $F_{er}$  are small, and so, backmixing has essentially no effect.

TABLE 2. Comparison of conversions calculated and those observed

Run No.	$\eta$ obsd.	$\eta$ calcd.	
		P·P-type	P·M-type
13	0.26	0.219	0.173
14	0.14	0.105	0.077
15	0.10	0.073	0.057

In order to assume the approximate values of  $E_z$  used in the calculation of Table 1, Equations (36)~(38) obtained by the author *et al.*<sup>11)</sup> were adopted.

When  $Re_b/(1-\epsilon) = 15 \sim 50$

$$E_z/\nu = 100\{Re_b/(1-\epsilon)\}^{0.95} \quad (36)$$

When  $Re_b/(1-\epsilon) = 50 \sim 200$

$$E_z/\nu = 180\{Re_b/(1-\epsilon)\}^{0.8} \quad (37)$$

$$\epsilon = \epsilon_c(u_0/u_{mf})^{5.7 D_p} \quad (D_p: \text{cm}) \quad (38)$$

Height of fluidized bed can be estimated by combining Equations (39) with (38).

$$L_f = L_c(1-\epsilon_c)/(1-\epsilon) \quad (39)$$



The value of  $u_{mf}$  adopted in these equations was evaluated by means of the empirical equation obtained by Leva *et al.*<sup>12)</sup>

Calculating by P•P-type parallel-flow reactor model, the distribution of concentration in each phase and the variation of average concentration are illustrated in Fig. 6. It is seen that average concentration at the top of bed does not become equal to the outlet concentration, and such disagreement is due to the difference of definitions which the former is the average based on sectional area and the latter is based on flow-rate<sup>13)</sup>.

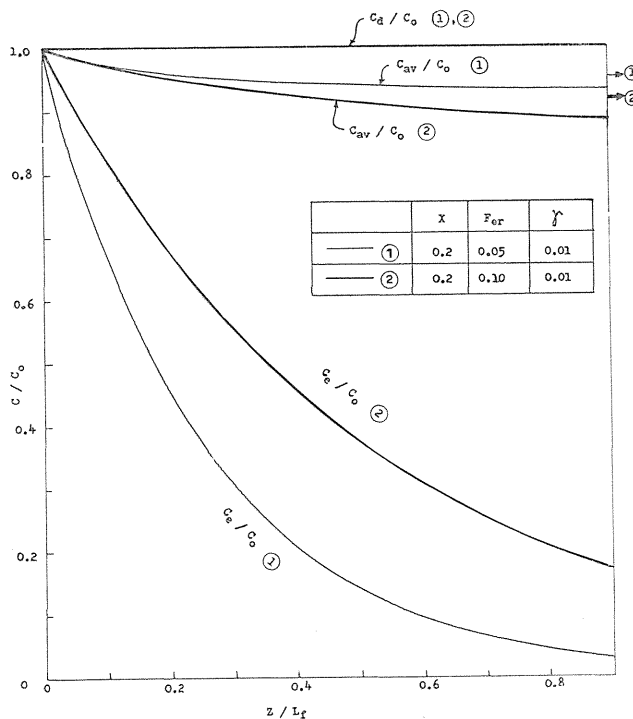


FIG. 6. Concentration distributions in P•P-type parallel-flow model

**Conclusion**

In order to determine the over-all conversion in fluidized-bed catalytic reactor, Equation (14) of inclusive two-phase model was derived. And it was shown that various two-phase models and homogeneous phase model could be derived from Equation (14).

Effect of  $E_z$  on conversion in two-phase model was clarified graphically in Fig. 2 and 3.

**Acknowledgement**

The author wishes to thank Mr. I. Yamada and T. Mamuro for assistance in analysing the computer data.

## Nomenclature

$A_e$	: effective cross-sectional area for gas flow in emulsion phase [cm <sup>2</sup> ]
$A_r$	: cross-sectional area of fluidized-bed reactor [cm <sup>2</sup> ]
$a_i$	: effective interfacial area between dilute and emulsion phases per unit volume of bed [cm <sup>2</sup> /cm <sup>3</sup> ]
$C_d, C_e$	: concentration of reactant in dilute and emulsion phases, respectively [mol/cm <sup>3</sup> ]
$C_{d1}, C_{e1}$	: concentration of reactant at the top of dilute and emulsion phases, respectively [mol/cm <sup>3</sup> ]
$C_0, C_1$	: concentration of reactant at the inlet and outlet of fluidized-bed reactor, respectively [mol/cm <sup>3</sup> ]
$C_e^0$	: concentration of reactant at the bottom of emulsion phase [mol/cm <sup>3</sup> ]
$D_p$	: particle diameter [cm]
$E_z$	: longitudinal-dispersion coefficient of gas [cm <sup>2</sup> /sec]
$F_c$	: gas-interchange rate per unit interfacial area [cm <sup>3</sup> /cm <sup>2</sup> ·sec]
$F_d, F_e$	: volumetric gas flow rate through dilute and emulsion phases, respectively [cm <sup>3</sup> /sec]
$F_{dr}, F_{er}$	: ratio of $F_d$ and $F_e$ to $F_t$ , respectively [-]
$F_t$	: volumetric flow rate of total gas in fluidized-bed reactor [cm <sup>3</sup> /sec]
$k$	: 1st-order reaction rate constant [cm <sup>3</sup> /g(cat.)·sec]
$L_c$	: packed bed height [cm]
$L_{mf}$	: bed height at minimum fluidization [cm]
$L_f$	: fluidized-bed height [cm]
$Re_p$	: $D_p u_0 \rho / \mu$ [-]
$U$	= $u L_f / 2 E_z$ [-]
$U_e$	= $u_e L_f / 2 E_z$ [-]
$u$	: average gas velocity in particulate fluidized-bed [cm/sec]
$u_e$	: average gas velocity in emulsion phase [cm/sec]
$u_0$	: superficial gas velocity [cm/sec]
$u_{mf}$	: superficial minimum fluidization velocity [cm/sec]
$W_s$	: total mass of catalyst in static-bed reactor [g]
$X$	: $k W_s / F_t$ [-]
$z$	: longitudinal distance from bottom of bed [cm]
$\gamma$	: catalyst fraction in dilute phase [-]
$\varepsilon$	: average fractional void in particulate fluidized-bed [-]
$\varepsilon_c$	: average fractional void in static bed [-]
$\varepsilon_{mf}$	: average fractional void at minimum fluidization [-]
$\mu$	: viscosity of fluid [g/cm·sec]
$\nu$	: kinematic viscosity [cm <sup>2</sup> /sec]
$\rho$	: density of fluid [g/cm <sup>3</sup> ]
$\eta$	: conversion [-]

## References

- 1) Kobayashi, H. and F. Arai: Preprint of 1st Anniversary Symposium (The Soc. of Chem. Engrs., Japan), pp. 201 (1962).
- 2) May, W. G.: Chem. Eng. Progr., **55**, No. 12, 49 (1959).
- 3) Danckwerts, P. V.: Chem. Eng. Sci., **2**, 1 (1953).

- 4) Yagi, S. and T. Miyauchi: *Kagaku-Kogaku* (Chem. Eng., Japan), **19**, 507 (1955).
- 5) Towe, R., R. Matsuno, H. Ishii, and H. Kojima: Preprint of 3rd. Anniversary Symposium (The Soc. of Chem. Engrs., Japan), pp. 32 (1964).
- 6) Davidson, J. F. and D. Harrison: "Fluidized Particles", pp. 100 (Cambridge Univ. Press). (1963).
- 7) Massimilla, L., H. F. Johnstone: *Chem. Eng. Sci.*, **16**, 105 (1961).
- 8) Shen, C. Y. and H. F. Johnstone: *A.I.Ch.E. Journal*, **1**, 349 (1955).
- 9) Johnstone, H. F., J. D. Batchelor and C. Y. Shen: *A.I.Ch.E. Journal*, **1**, 318 (1955).
- 10) Mathis, S. F. and C. C. Watson: *A.I.Ch.E. Journal*, **2**, 518 (1956).
- 11) Muchi, I., T. Mamuro and K. Sasaki: *Kagaku-Kogaku* (Chem. Eng., Japan), **25**, 747 (1961).
- 12) Leva, M., T. Shirai and C. Y. Wen: *Genie Chimique*, **75**, 33 (1956).
- 13) Muchi, I., T. Mamuro and S. Mori: Preprint of 30th. Anniversary Meeting (The Soc. of Chem. Engrs., Japan), pp. 453 (1965).