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Combined the authors have accumulated more than two decades of industrial experiences in the process control and reaction kinetic modeling of petroleum processes. They report that their efforts in the process control area have been successful and rewarding, but tempered with the occasional blunting of their swords in the implementation of advanced control strategies, the latter of which is emphasized intentionally in the paper to expose some of their anxieties regarding advanced control techniques. The present paper was originally prepared as a plenary lecture at the 1974 JACC.

Manuscript received July 10, 1975, and accepted November 3, 1975.

Experimental Investigation of Models for Fluidized Bed Catalytic Reactors

The countercurrent backmixing model of a fluidized bed reactor predicts axial concentration profiles quite different from those suggested by simple two-phase models. The models can also be distinguished in terms of the dependence of conversion on operating variables.

An experimental study of ozone decomposition in a reactor of 22.9 cm diameter has provided extensive data for comparison with backmixing and two-phase models which incorporate bubble size variation. The measured profiles show a minimum concentration within the bed at gas velocities above a critical value, as predicted only by the backmixing model. The effect of operating variables on the shape of the profiles is also well accounted for by this model. The backmixing model is further supported by good agreement between predicted and measured reactant conversion. In particular, the variation of conversion with rate constant and gas velocity is fitted more accurately by the backmixing model than by two-phase models.

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SCOPE

The countercurrent backmixing model, proposed by several investigators, attempts to account for gas mixing behavior in bubbling gas-fluidized beds of fine particles. The model predicts downflow of gas with solids in the particulate phase at fluidizing gas velocities above some critical value. The earlier and simpler two-phase models of Davidson and Harrison (1963) assume either that gas

in the particulate phase is completely mixed or that gas flows upward in plug flow through the particulate phase.

Previous experimental studies have shown that the backmixing model provides a good description of mixing of a tracer gas injected just under the surface of a fluidized bed. Available data on fluidized bed reactor performance are not suitable for testing the validity of the

backmixing model; bubble sizes are rarely reported, experiments have been conducted at conditions where predictions of various models are not very different, and hardly any investigators have measured reactant concentration profiles within the bed.

Fryer and Potter (1972a) showed that the backmixing model differs significantly from the two-phase models in predicting the dependence of conversion on rate constant and on gas velocity in a catalytic reactor. Furthermore, they pointed out a unique result of the backmixing

model, reactant concentration profiles show a minimum value at a position within the bed. These conclusions opened the way for the experimental program designed specifically to test the backmixing model in a catalytic reactor, which is reported in this paper. To allow a valid comparison of experimental results and model predictions, all model parameters have been experimentally determined and all models have been modified to incorporate a description of bubble size variation.

CONCLUSIONS AND SIGNIFICANCE

The two-phase and backmixing models have been compared with experimental results from a study of the catalytic decomposition of ozone on sand particles of mean diameter $117\mu\text{m}$ in a reactor of 22.9 cm diameter. Bed expansion, bubble diameter, bubble frequency, conversion, and axial concentration profiles have been measured over wide ranges of fluidizing gas velocity, bed height, and reaction rate constant. Bubble diameter varies linearly with height up to a size of about 10 cm equivalent diameter; beyond this size the models under consideration could not be applicable because of slugging behavior.

The shapes of the observed axial concentration profiles are as predicted only by the backmixing model, with an apparent critical gas velocity of the order of 5 cm s^{-1} . At velocities below this, the ozone concentration decreases gradually from distributor to bed surface but at higher velocities passes through a minimum value at a position within the bed. This value of critical velocity

indicates a volume ratio of wake to bubble of the order of 0.8. The effect of operating variables on the position of the minimum in the concentration profiles is also well predicted by the backmixing model.

Further support for the backmixing model is provided by the conversion data, which are in close agreement with model predictions. In particular, the variation of conversion with rate constant and gas velocity is predicted more accurately by the backmixing model than by the two-phase models.

It is concluded that the backmixing model in the form presented by Fryer and Potter (1974) provides a good description of the reactor behavior. Simple two-phase models cannot account for the observed shape of the concentration profiles. Further research is required to study the effects of gas adsorption and radial nonuniformity, to verify the theoretical equations used for gas exchange, and to provide reliable means for predicting wake size and variation of bubble size with height.

Some early results of an experimental study of the validity of the backmixing model of a gas-fluidized reactor have already been reported (Fryer and Potter, 1974). That study has now been concluded (Fryer, 1974); this paper presents a comparison of experimental results with the backmixing model and with various forms of the two-phase models of Davidson and Harrison (1963).

Davidson and Harrison (1963) presented two two-phase models of fluidized beds. In one, the gas flow through the dense phase is in plug flow; in the other, the dense phase gas is completely mixed. In both models, gas exchange between phases is written as

$$\frac{Q}{V} = \frac{4.5 U_{mf}}{D} + \frac{5.85 D_G^{0.5} g^{0.25}}{D^{1.25}} \quad (1)$$

This equation is based on diffusive transfer from a bubble and convective transfer from a bubble in a fluidized bed to the surrounding cloud, but it was used by Davidson and Harrison (1963) for transfer to the entire surrounding dense phase. It seems more consistent (Potter, 1971) to replace Q/V by K_{BP} , the overall exchange coefficient resulting from the two-step exchange process proposed by Kunii and Levenspiel (1968):

$$\frac{1}{K_{PB}} = \frac{1}{K_{BC}} + \frac{1}{K_{CP}} \quad (2)$$

where

$$K_{BC} = \frac{Q}{V} \quad (\text{bubble to cloud transfer}) \quad (3)$$

$$K_{CP} = 6.78(\epsilon_{mf} D_G u_A D^{-3})^{0.5} \quad (\text{cloud to dense phase}) \quad (4)$$

The backmixing model first advanced by Stephens et al. (1967) and further elaborated by Lathem et al. (1968) and, independently, by Kunii and Levenspiel (1968) who used the description "bubbling-bed model" attempts to provide a more satisfactory description of gas mixing in fluidized beds. The rate of downflow of solids in the dense phase is equated to the upflow in the bubble wakes, and it is assumed that the relative velocity of gas to solids in the dense phase is the same as that at incipient fluidization. Thus the model predicts downflow of gas with solids in the dense phase at gas flow rates above some critical value. The various early simplified forms of this model were considered by Fryer and Potter (1972a), who concluded that for a reaction system only a rigorous solution of the three-phase model (with bubble, cloud-wake, and particulate phases) is satisfactory over a reasonable range of operating variables. They retained two assumptions, namely, that the volume of the bubble cloud is negligible and that there are no particles in the bubble phase. Fryer (1974) has shown that these assumptions have an insignificant effect on catalytic reactor model predictions.

It has been pointed out (Fryer and Potter, 1972b) that it is necessary to incorporate bubble size variation into fluidized bed reactor models before any worthwhile comparison with experimental results can be made. Solutions have been presented for two-phase models (Fryer and Potter, 1972b) and for the three-phase countercurrent backmixing model (Fryer and Potter, 1974) with linear bubble size variation, following Kato and Wen (1969)

$$D = D_0 + mh \quad (5)$$

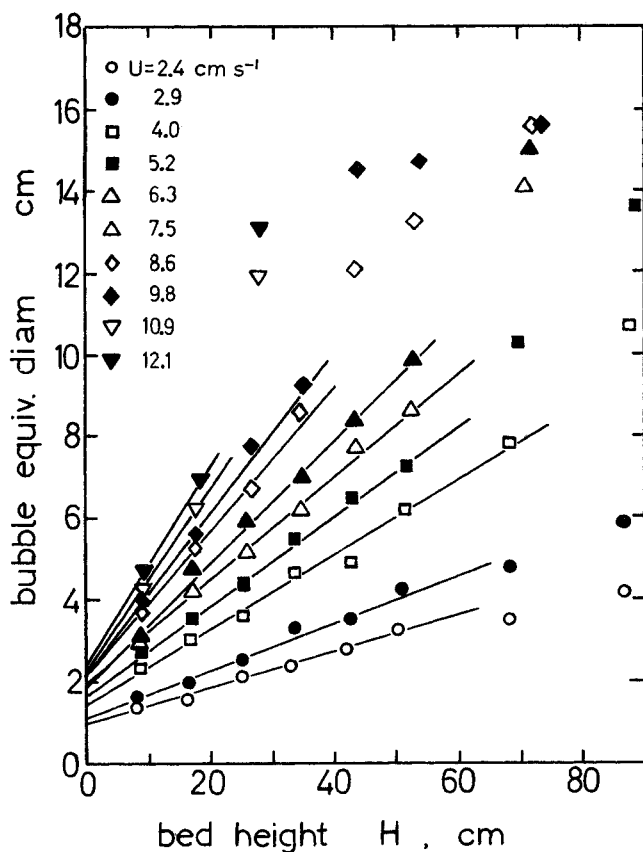


Fig. 1. Variation of bubble size with height.

and Kobayashi et al. (1966)

$$m = 1.4 d_p \rho_s \left(\frac{U}{U_{mf}} \right) \quad (6)$$

The experimental program reported here was designed to provide comprehensive data on reactor conversions and concentration profiles at conditions most favorable for distinguishing between the various forms of the simple two-phase models and the backmixing model. All necessary model parameters, including bubble sizes, have been experimentally determined.

EXPERIMENTAL EQUIPMENT AND CATALYST

The experimental apparatus has already been described (Fryer and Potter, 1974) and specified in detail (Fryer, 1974). It consists of a 200 cm × 22.9 cm diameter stainless steel and glass reactor with a bubble cap distributor (a plate fitted with 61 caps on 27.8 mm spacing; each cap is 18 mm high × 9 mm diameter with four 0.8 mm diameter holes). A recirculating dry air system supplies the fluidizing gas; ozone is produced in an electrical-discharge ozonator and added with diluent air to the circulating gas at the inlet of the reactor. Through the top cover plate of the reactor pass thirteen gas sample probes which can be moved vertically. One is positioned on the central axis, the others on two perpendicular diameters at 3.3 cm spacings. Each probe is of stainless steel tubing, 7.1 mm internal diameter, fitted with a sintered glass disk over the end.

The ozone decomposition reaction under investigation is carried out in the reactor bed at room temperature on a catalyst of sand impregnated with iron oxide ($d_p = 117 \mu\text{m}$, $U_{mf} = 1.70 \text{ cm s}^{-1}$, $\epsilon_{mf} = 0.48$, $\rho_s = 2.65 \text{ g cm}^{-3}$). The catalyst is protected from poisons (for example, from the recirculating blower) by passing the incoming gas through a bed of molecular sieve pellets.

Gas samples from the reactor inlet and outlet gas streams, and from internal probes, are analyzed for ozone concentration by differential absorption of ultraviolet light in a continuous flow meter. The frequency of bubbles and their mean diameter

have been measured from movie film of bubbles erupting at the surface of beds of various heights. Previous experience in fluidized beds of 15 cm diameter, fitted with capacitance bands to detect bubbles, has shown that such data provide satisfactory information on bubble frequencies within fluidized beds; that is, static head of bed above the bubble has no detectable effect.

In parallel with the fluid bed reactor is a glass fixed bed reactor of 2.64 cm diameter and 61 cm length. This has been used for preliminary testing of the catalyst and to obtain reaction rate constants on catalyst samples taken from the fluidized bed. A gas sample stream can be taken from the outlet of the fixed bed for analysis in the ozone meter.

EXPERIMENTAL PROGRAM

Bubble Sizes

For each bubble eruption filmed, the eruption diameter was measured as the mean of two bubble widths at right angles. For each set of conditions (that is, bed height and gas velocity), forty to forty-five bubbles were measured and the arithmetic mean eruption diameter calculated. Conversion to volumetric equivalent diameter D has been based on the conclusion of Whitehead and Young (1967)

$$D = 0.78 \times \text{eruption diameter} \quad (7)$$

The equivalent diameters thus calculated are plotted as a function of bed height in Figure 1 to test the validity of Equation (5). The data conform quite well to the linear form of Equation (5), except for results where the equivalent diameters exceed 10 cm (eruption diameters greater

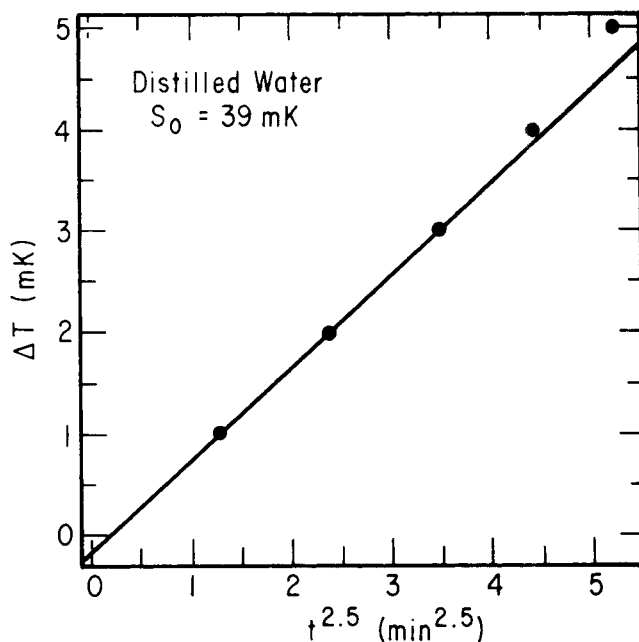


Fig. 2. Dependence of slope m on fluidizing gas velocity U .

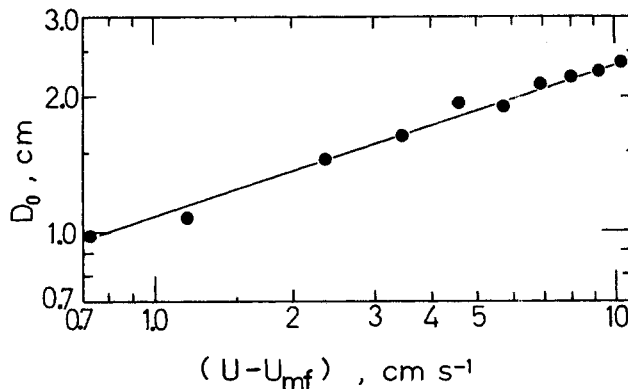


Fig. 3. Dependence of D_0 on $(U - U_{mf})$.

TABLE 1. ACTIVITY OF CATALYST SAMPLES

| | | A = Sample withdrawn early in run B = Sample withdrawn late in run | | | |
|-------------------------|-------------|---|--------------------------|-----------------------|----------------------------|
| Catalyst activity level | Sample time | Fixed bed ht, cm | U , cm s ⁻¹ | k , s ⁻¹ | Mean k , s ⁻¹ |
| 1 | A | 3.7 | 15.2 | 7.73 | 7.75 |
| | A | 3.7 | 27.1 | 8.26 | |
| | B | 3.7 | 15.2 | 7.05 | |
| | B | 3.7 | 27.2 | 7.96 | |
| 2 | A | 3.8 | 15.3 | 3.57 | 3.60 |
| | A | 3.8 | 27.4 | 4.28 | |
| | B | 3.7 | 15.2 | 3.20 | |
| | B | 3.7 | 26.9 | 3.34 | |
| 3 | A | 7.7 | 20.7 | 1.51 | 1.57 |
| | A | 7.7 | 12.2 | 1.63 | |
| 4 | A | 7.7 | 20.8 | 0.88 | 0.86 |
| | A | 7.7 | 13.8 | 0.84 | |
| 5 | A | 11.6 | 15.1 | 0.34 | 0.33 |
| | A | 11.6 | 11.2 | 0.33 | |
| | B | 11.2 | 15.7 | 0.32 | |
| | B | 11.2 | 8.8 | 0.33 | |
| 6 | A | 15.6 | 13.1 | 0.14 | 0.14 |
| | A | 15.6 | 8.0 | 0.15 | |
| | B | 15.1 | 13.2 | 0.14 | |
| | B | 15.1 | 8.8 | 0.14 | |
| 7 | A | 21.6 | 10.7 | 0.040 | 0.049 |
| | A | 21.6 | 7.1 | 0.059 | |
| | B | 31.6 | 8.1 | 0.043 | |
| | B | 31.6 | 6.4 | 0.054 | |

than 13 cm). At these conditions in the 22.9 cm diameter reactor, the bed is entering a slugging regime. Equation (7) will no longer be valid, owing to change in bubble shape, and the bubbling bed models, which assume radial uniformity, cannot be expected to apply. Therefore, comparison of reactor performance and model predictions will not be made under these conditions.

Figure 2 shows the dependence of the slope m in Figure 1 on fluidizing velocity. The data are well fitted by a straight line through the origin, consistent with Equation (6). The slope of the line of best fit in Figure 2 is a little lower than that predicted by Equation (6), which is based on bubble height rather than on equivalent diameter. The experimental line

$$m = 2.05 \times 10^{-2} U \quad (\text{in c.g.s. units}) \quad (8)$$

will be used in computing model predictions.

Extrapolation in Figure 2 allows determination of D_o , the bubble size at the distributor, for each gas velocity. From the theory of bubble formation (Harrison and Leung, 1961), it is expected that the bubble volume at the distributor should be proportional to $(U - U_{mf})^{1.2}$, that is, D_o should be proportional to $(U - U_{mf})^{0.4}$. Figure 3 shows that the data are satisfactorily described by

$$D_o = 1.08(U - U_{mf})^{0.33} \quad (\text{in c.g.s. units}) \quad (9)$$

and this relation will be used in computing model predictions. The description of bubble history provided by Equations (5), (8), and (9) is likely to be applicable only to the equipment used in this study. In particular, the bubble size at the distributor would differ from that given by Equation (9) if different types of distributor were used.

In addition to the bubble size data discussed above, bubble frequencies have been measured. By assuming that the apparent superficial velocity of bubble gas U_{GB} is equal to $(U - U_{mf})$, an equivalent diameter can be

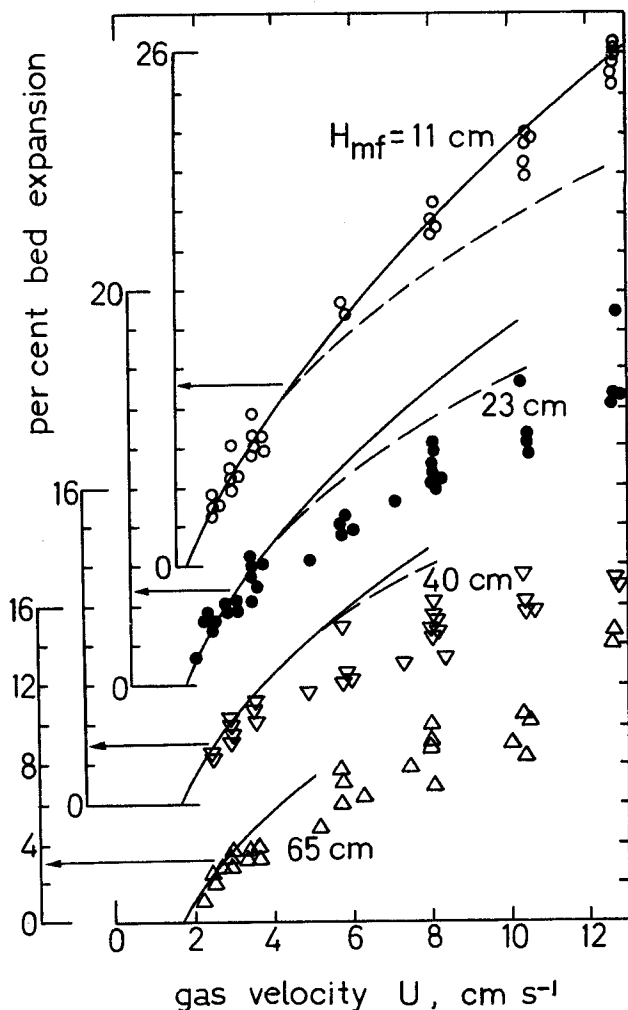


Fig. 4. Bed expansion compared with model predictions.
— predictions using measured bubble diameters.
--- predictions using diameters calculated from frequency data.

calculated for each set of conditions. This calculation shows ratios of equivalent diameter to measured eruption diameter varying from 0.65 to 0.98 [0.78 in Equation (7)]. This apparent discrepancy may be due either to departure of the value of U_{GB} from $(U - U_{mf})$ or to variations in bubble shape. Diameters derived from the frequency data differ slightly from those predicted by Equations (8) and (9), but, although the model is sensitive to bubble diameter, the differences are not large enough to alter the model predictions significantly (Fryer, 1974).

Catalyst Activity

Preliminary tests in the fixed bed reactor confirmed that ozone decomposition is close to first order, whether carried out on the original quarry sand of wide size distribution, on the -100 + 150 mesh portion (117 μ m mean diameter) prepared for use in the fluidized bed reactor studies, or on the 117 μ m material impregnated with iron oxide as described by Fryer and Potter (1974).

During the operation of the fluidized bed reactor, small samples of catalyst particles were taken from the fluidized reactor and each tested at two gas velocities in the fixed bed reactor. Samples were taken at each of the seven levels of catalyst activity used in the reactor studies. The results, in Table 1, confirm that the reaction is close to first order, since there is good agreement in values of the rate constant k calculated at different velocities for each level of activity. There is no significant difference

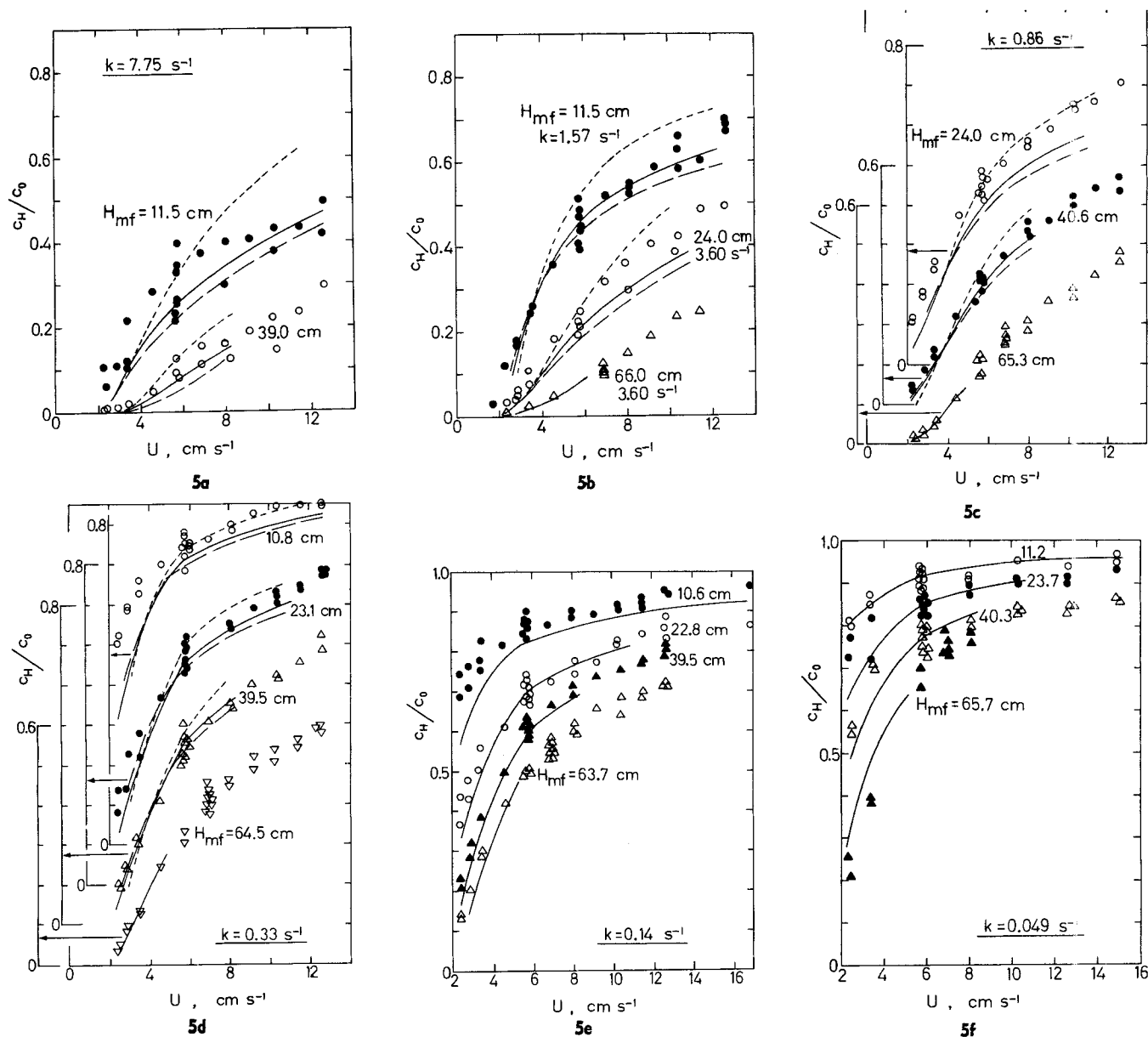


Fig. 5. Conversion data compared with predictions of backmixing model (variation of U).

— model with measured bubble diameters, $f_w = 0.75$.
 --- model with measured bubble diameters, $f_w = 1.0$.
 - · - model with diameters from frequency data, $f_w = 0.75$.

between the activities of samples taken near the start (A in Table 1) and near the end (B) of the experimental runs. The mean values of k will be used in computation of model predictions for comparison with the experimental fluidized bed reactor results.

The Fluidized Bed Reactor

Blank runs were carried out with no catalyst in the fluidized bed nor in the small fixed bed reactor to check that all gas samples showed the same ozone concentration under this condition. Only the samples from the reactor probes showed variation, with a loss of 3 to 13% of ozone on passing through the tip and tube of the probe. Each probe was calibrated by plotting the registered concentration of the probe sample against that of the reactor outlet gas. During later reaction runs these calibrations were checked at the beginning and end of each recorded axial profile.

For each level of catalyst activity, the fluidized bed reactor was first charged with approximately 80 lb (36.3 kg) of catalyst, reduced subsequently by discharge of

weighed quantities for study of lower beds (down to 15 lb). Some of the catalyst materials of lower activity showed a slight tendency to increase in activity during use. This was controlled by use of a small bypass around the molecular sieve on the gas inlet.

In all, catalysts with seven activities have been studied (see Table 1) at gas velocities ranging from 2.4 to 17.3 cm s^{-1} . At all 293 sets of conditions, ozone concentrations in the inlet and outlet gas were recorded, providing conversion data for comparison with model predictions. At 106 selected conditions, the probes were used to record axial profiles within the bed and to measure the expanded bed height. In early reaction runs, it was shown that ozone concentration was very close to symmetrical about the central axis of the bed; thus axial profiles taken with the central probe and three other probes across one radius are sufficient to describe bed behavior. This assumption of symmetry was checked in detail at six widely different sets of conditions during the course of the reaction runs.

COMPARISON OF EXPERIMENTAL RESULTS WITH MODEL PREDICTIONS

Values of Model Parameters

Experimental values of bed expansion, conversion, and concentration profiles are to be compared with predictions of the two-phase models (Fryer and Potter, 1972b) and the backmixing model (Fryer and Potter, 1974) with bubble size variation. In the model computations, experimentally determined values of U , H_{mf} , U_{mf} (1.70 cm s^{-1}), ϵ_{mf} (0.48), k (see Table 1), and bubble size parameters D_0 and m [see Equations (8) and (9)] have been used. The diffusivity of ozone in air, D_G , at a temperature of 20° to 25°C , has been taken as $0.18 \text{ cm}^2\text{s}^{-1}$ by using data from Davidson and Harrison (1963) and the temperature dependence recommended by Perry (1950).

The only other parameter required is f_w , the ratio of wake volume to bubble void volume. Its value can be estimated from inspection of the measured ozone concentration profiles. All profiles recorded at gas velocities of 5.8 cm s^{-1} and above show a minimum concentration within the bed (see presentation and further discussion of profiles below). As has been widely discussed (Latham et al., 1968; Fryer and Potter, 1972a, 1974), this is characteristic of backmixing behavior. It provides immediate evidence in support of the backmixing model and indicates that for the catalyst particles used in this work, the critical fluidizing velocity U_{cr} is less than 5.8 cm s^{-1} . On the other hand, all profiles at gas velocities of 3.5 cm s^{-1} and below show a gradual decline in concentration from the distributor right to the surface. A number of checks at a gas velocity of 4.6 cm s^{-1} revealed no inversion of ozone concentration, so U_{cr} lies in the range 4.6 to 5.8 cm s^{-1} . As derived by Fryer and Potter (1972a, 1974)

$$\frac{U_{cr}}{U_{mf}} = \left[1 + \frac{1}{\epsilon_{mf} f_w} \right] [1 - \epsilon_B (1 + f_w)] \quad (10)$$

Expansion data indicate that ϵ_B is about 0.04 at $U = 4.6 \text{ cm s}^{-1}$ and about 0.06 at $U = 5.8 \text{ cm s}^{-1}$. Equation (10) then shows f_w to be in the range 1.02 to 0.75, and the experimental indications are that it is probably closer to the lower value. For a discussion of pertinent data relating to f_w see Potter (1971).

Bed Expansion

In Figure 4, experimental expansion data are compared with predictions from the models incorporating a linear variation of bubble size with height. The comparison is restricted to those conditions at which bubble sizes are low enough for the models to be valid, as discussed earlier.

Agreement between predicted and observed expansion is good, especially for the lower bed heights. The small discrepancies for higher beds are not surprising, considering the difficulty of measurement caused by vigorous motion of the surface. The method of measurement, involving observation of a probe tip from above, may well underestimate the average bed height. Overall, it is clear that the description of bubble rise velocity used in the models is adequate, leading to a reasonably good prediction of expanded bed height.

Conversion—Comparison of Backmixing Model with Experiment

Figure 5 shows conversion data as dimensionless outlet concentration c_H/c_0 as a function of fluidizing velocity at all seven levels of catalyst activity. The corresponding predictions of the backmixing model of Fryer and Potter (1974) have been computed with $f_w = 0.75$. At some conditions, model predictions with $f_w = 1.0$ and predic-

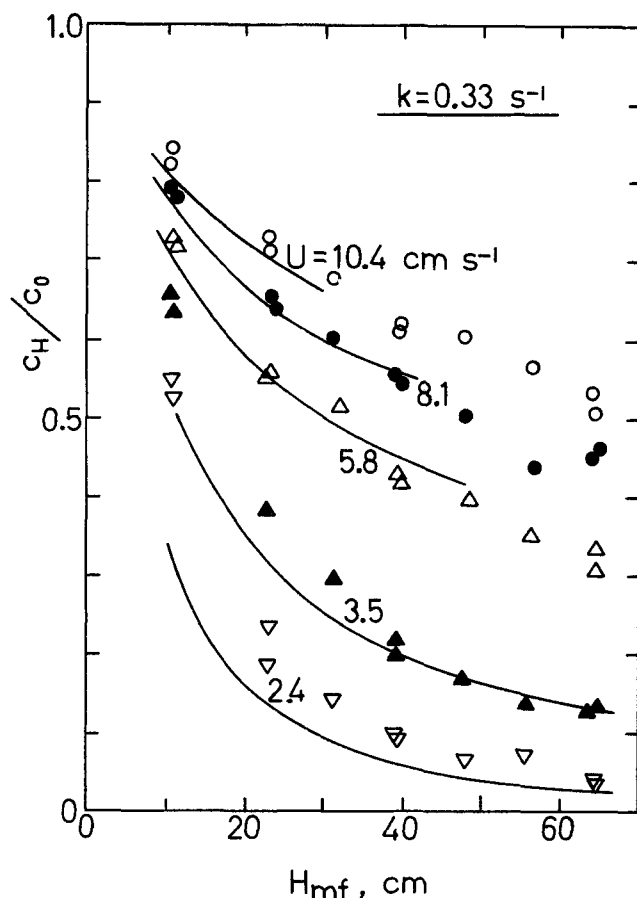


Fig. 6. Conversion data compared with predictions of backmixing model (variation of H_{mf}).

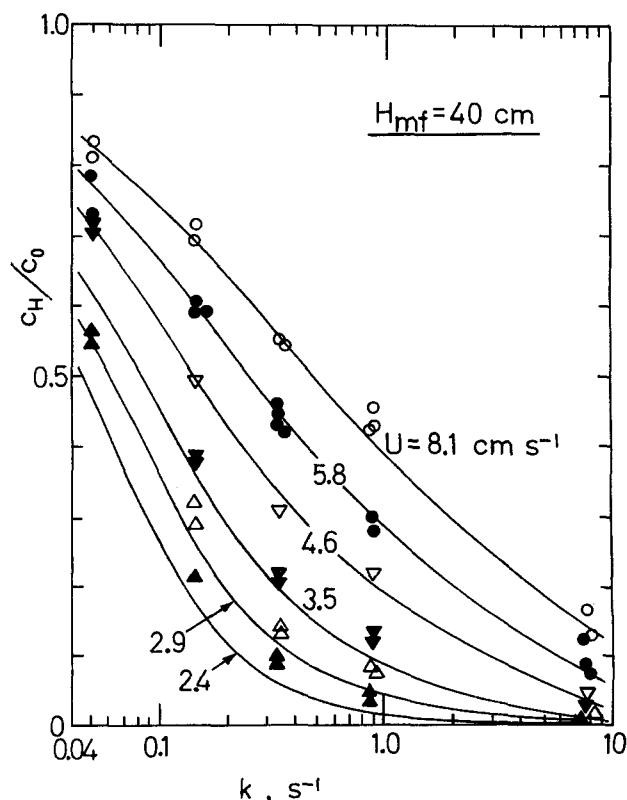


Fig. 7. Conversion data compared with predictions of backmixing model (variation of k).

tions using bubble diameters computed from frequency data are also shown.

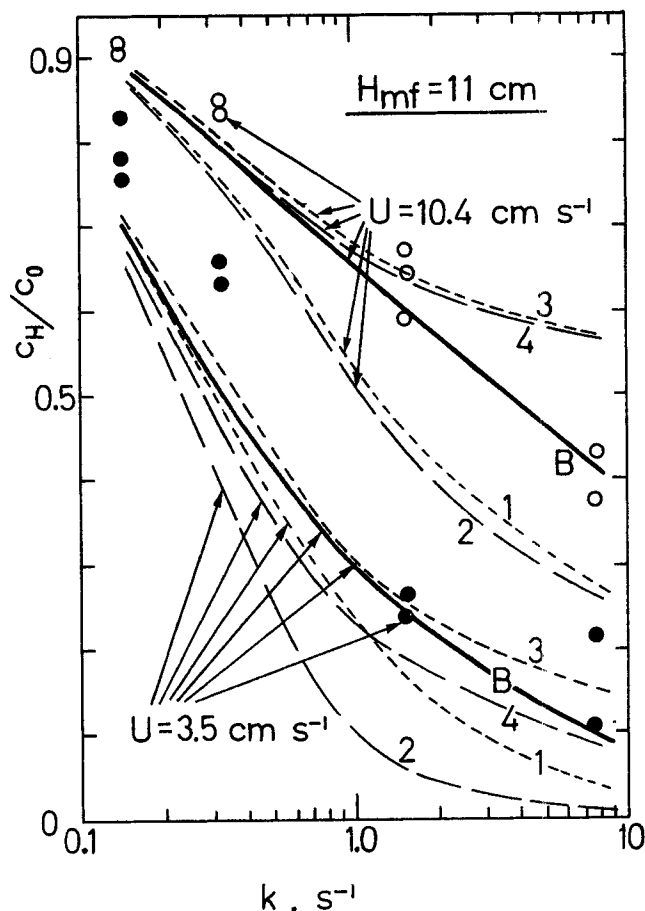
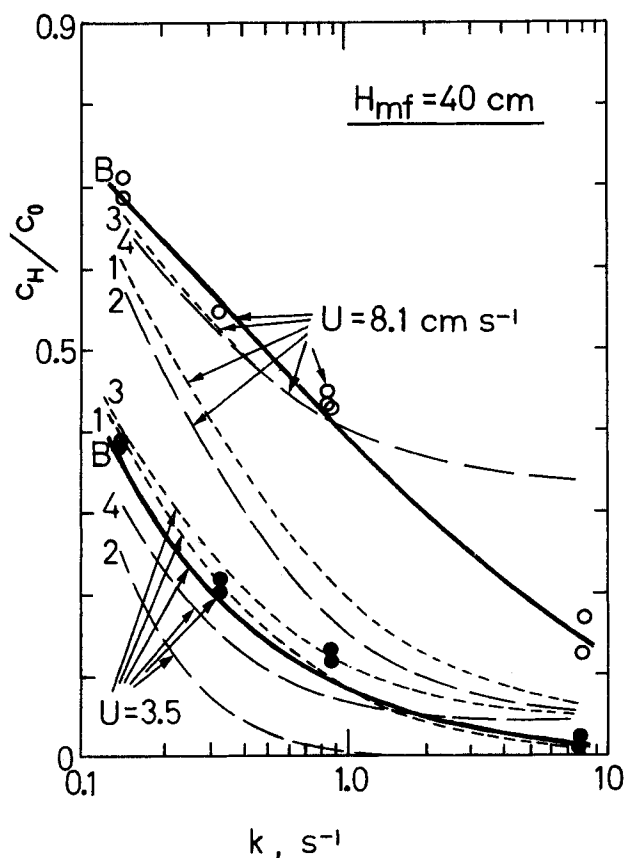


Fig. 8. Comparison of models with experimental conversion data (variation of k).

- B ——— backmixing model.
- 1 - - - - two-phase model, well-mixed dense phase gas, Davidson exchange.
- 2 - - - - two-phase model, plug flow dense phase gas, Davidson exchange.
- 3 - - - - two-phase model, well-mixed dense phase gas, Kunii exchange.
- 4 - - - - two-phase model, plug flow dense phase gas, Kusii exchange.

The experimental data are in good agreement with model predictions at all conditions. In particular, the data clearly follow the distinctive change of slope in the vicinity of the critical fluidizing velocity, as predicted only by the backmixing model.

For clarity, data at some values of H_{mf} have been omitted from Figure 5. Figure 6 shows c_H/c_0 vs. H_{mf} , allowing inclusion of additional data at $k = 0.33 \text{ s}^{-1}$ and providing a check on whether the backmixing model correctly predicts the dependence of conversion on bed height. Generally, agreement is again satisfactory, but the observed outlet concentrations are somewhat higher than predicted at low values of H_{mf} and U . The same trend is observed with data at other values of k . This discrepancy may be due to the fact that the gas enters the bed 1.3 cm above the plate, passing through a smaller depth of catalyst than accounted for in the model. This would be most significant at low values of H_{mf} , and also at low values of U , when a greater proportion of gas passes through the particulate phase. Furthermore, portions of the bed above and between the caps may be inactive at low gas velocities. Note that the model assumes the smallest bubble (and most rapid gas exchange) in these lowest parts of the bed, where there may be no gas flow at all.

Finally, it is of interest to examine the data in terms of the dependence of conversion on catalyst activity. Typical results are plotted in Figure 7, again showing that the backmixing model provides a good description of reactor performance, with a tendency to predict outlet concentrations which are a little too high at low values of gas velocity.

Conversion—Comparison of Backmixing and Two-Phase Models

The model comparisons of Fryer and Potter (1972a) suggested that, in terms of conversion, the dependence on reaction rate constant would provide the most definite distinction between the backmixing model and the two-phase models. Predictions of the various models and experimental data at a number of selected conditions are compared in Figures 8 and 9. All the model predictions are computed using Equations (8) and (9) to describe variation of bubble size.

The variation of c_H/c_0 with k is adequately described only by the backmixing model. As k increases, the two-phase models using K_{BP} tend to approach too soon a steady value of outlet concentration, and this steady value is considerably higher than that observed experimentally. Those using Q/V predict outlet concentrations lower than those observed, and also probably flatten out at too low a value of k .

Figure 9 shows typical results for the dependence of c_H/c_0 on U at moderate values of k , where the two-phase models with K_{BP} do predict conversions of the right order of magnitude. Here again the backmixing model appears to be the most satisfactory, fitting the experimental data closely with a distinct change of slope in the vicinity of the critical fluidizing velocity.

Consideration of the data as c_H/c_0 vs. H_{mf} also indicates in absolute terms that the backmixing model fits the experimental data best, but there is no distinctive difference between the trends predicted by the various models, so no further evidence is provided in support of any of the models.

Reactor Concentration Profiles

As reported by Fryer and Potter (1974), the ozone concentrations detected by four probes across any one radius do not differ significantly in comparison with axial variations. Also, axial variations detected by the probes at any one set of conditions all follow the same trend. The data from four probes have therefore been averaged, as explained by Fryer and Potter (1974), to give a single experimental profile.

The backmixing model, with $f_w = 0.75$, has been used to predict axial profiles of c_{avg}/c_0 , the ratio of sample gas to inlet gas concentration, for all 79 sets of conditions at which axial profiles have been measured and at which the bed is not entering a slugging regime. Figure 10 shows typical results, namely, those from fifteen sets of conditions using the nominal 30 lb catalyst charges ($H_{mf} = 22.8$ to 24.0 cm) at three levels of catalyst activity. The solid lines on the graphs are the backmixing model predictions. Also shown at $k = 0.33 \text{ s}^{-1}$ are the profiles predicted by the two forms of Davidson's two-phase model.

All of the experimental results display one feature which has already been discussed. At velocities of 5.8 cm s^{-1} and higher, each concentration profile passes through a minimum at a position within the bed, the concentration rising as we pass from that position to the bed surface. At all lower velocities, this is not observed; the concentration falls steadily from distributor to bed surface. Only the backmixing model predicts such behavior, so the very presence of these minima provides strong support for the concepts of this model.

Because of the well-recognized difficulty of removing accurate and representative gas samples from a fluidized bed through a probe, it may be argued that the observed minima are apparent rather than real, that is, arise because of sampling defects. For example, it may be suggested that when the probe is near the exit it draws bubble gas from just above the bed and gives a higher concentration. Three factors lead us to conclude that such is not the case. Firstly, minimum concentrations have been observed at quite low positions within the bed. In Figure 10 the minima occur some 3 to 5 cm below the bed surface, at which position the probe tip was certainly entirely below the bed surface at all times. Figure 11 shows other data from some more extreme cases with the minima 7 to 16

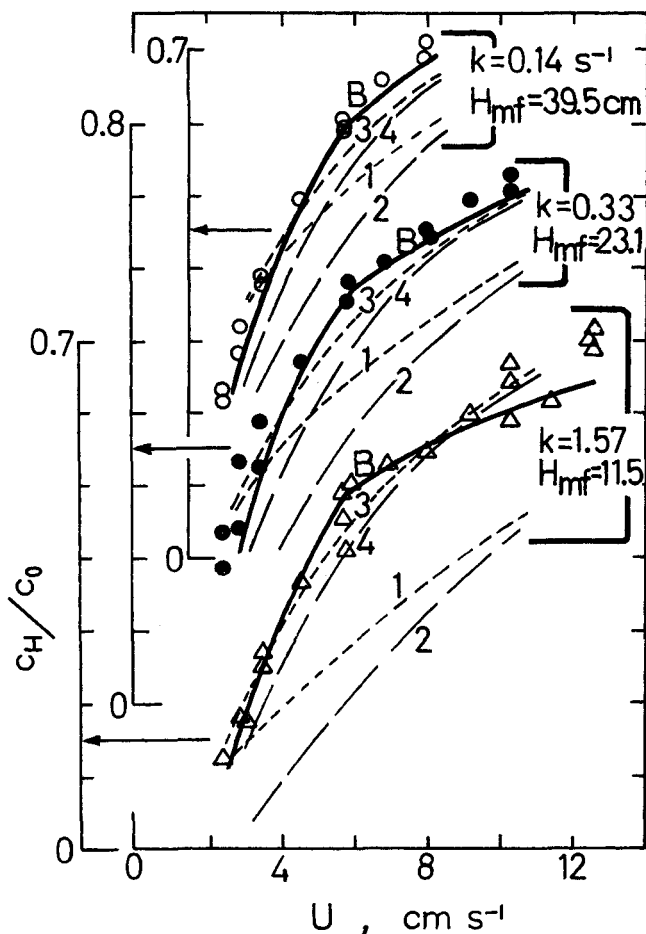


Fig. 9. Comparison of models with experimental conversion data (variation of U). (Models designated as in Fig. 8.)

cm below the surface. Secondly, the probe sample rate is equivalent to a gas velocity of only 0.9 cm s^{-1} across the cross-sectional area of the probe tip, so it is unlikely that gas would be pulled in from positions remote from the tip. Thirdly, any effect due to defective sampling would be more significant at low fluidizing gas velocities; minima which occur only when the fluidizing velocity exceeds some critical value are not likely to have arisen from such

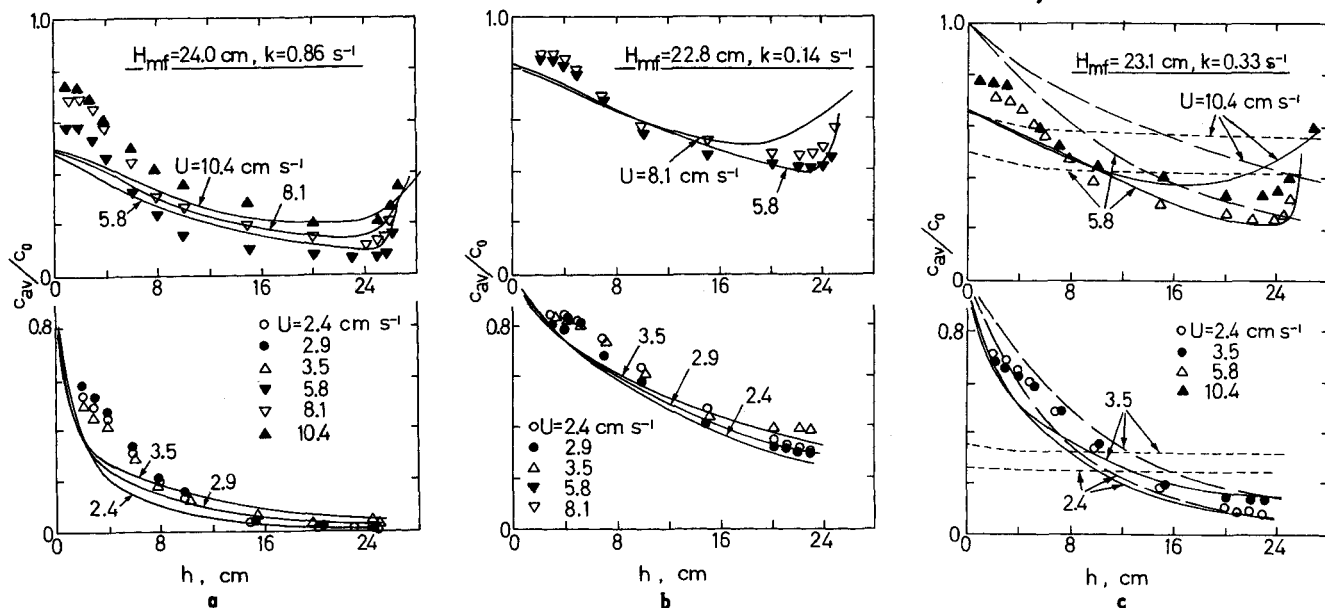
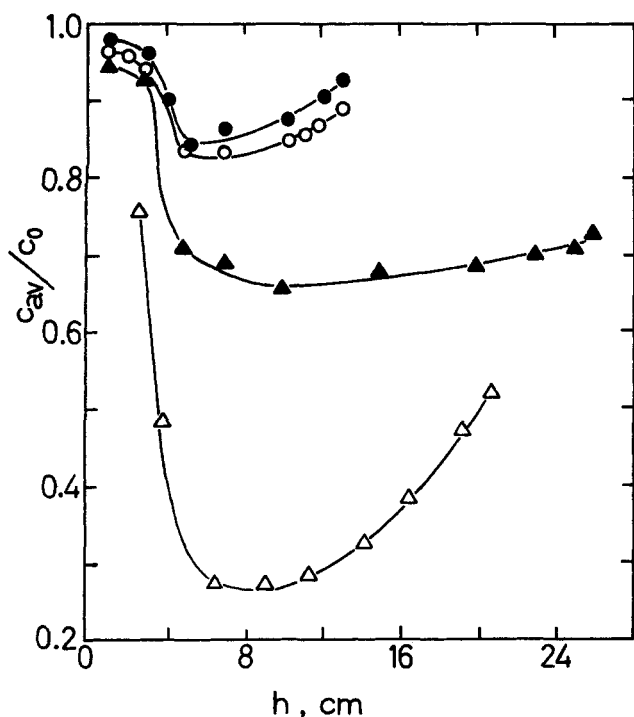


Fig. 10. Predicted and experimental profiles of reactant concentration.

— backmixing model.
 - - - two-phase model, well-mixed dense phase gas, Davidson exchange.
 - · - two-phase model, plug flow dense phase gas, Davidson exchange.



| | k, s^{-1} | H_{mf}, cm | $U, cm s^{-1}$ |
|---|-------------|--------------|----------------|
| ● | 0.049 | 11.2 | 15.0 |
| ○ | 0.14 | 10.6 | 12.7 |
| ▲ | 0.049 | 23.7 | 10.4 |
| △ | 0.10 | 20.2 | 8.5 |

Fig. 11. Experimental concentration profiles at selected conditions.

effects. It is interesting to note also that Schügerl (1974) has remarked that similar axial profiles with minimum concentrations were observed in investigation of hydrogenation of ethylene in fluidized beds.

Agreement between measured and predicted profiles is reasonably good, both in terms of the absolute values of c_{avg}/c_0 and in showing similar trends in the position of the minimum concentration (for $U > U_{cr}$); namely, the minimum occurs lower in the bed for higher values of U and for lower values of k , and lower in the bed, in proportion to total bed height, for lower values of H_{mf} . These trends are followed by all the experimental data.

As discussed earlier in relation to outlet concentration, departure from ideal conditions near the distributor plate causes measured concentration to be higher than predicted at conditions of low bed height and low gas velocity, and at low positions within the bed. It should also be noted that because meaningful measurements cannot be made below the tops of the distributor caps, it has not been possible to detect the predicted change in the shape of the profiles very close to the distributor when U exceeds U_{cr} . The backmixing model predicts that when $U < U_{cr}$, c_{avg}/c_0 will approach 1.0 near the distributor, but that under backmixing conditions c_{avg}/c_0 should level off at some lower value. The experimental data tend to show such a leveling off even at very low velocities.

CONCLUSIONS

The experimental data on reactant conversions and concentration profiles confirm the validity of the backmixing model. In the form considered here, it provides a promising description of fluidized bed reactor behavior. One must note, however, that the experimental evidence has been collected at conditions selected to minimize difficulties arising from two limitations of the model. Firstly,

the model takes no account of gross circulation within the bed, nor of any radial nonuniformity. Secondly, the effect of adsorption of reactant and product gases has been neglected. Further model development and experimental study are needed in both of these important areas.

The model is not tied to the gas-exchange equations used. It is to be expected that experiment will lead to more accurate values, although those employed here have done well in correlating the present data. What the model has achieved is the prediction that a concentration profile with a minimum concentration within the bed may occur, and this paper reports the experimental confirmation of this prediction. No other model has led its proponents to such a prediction.

In application of the backmixing model, two further problems arise. Data on the size of bubble wakes are sparse, and very little is known about the effect of bed variables on wake size. Secondly, it is not yet possible to predict bubble sizes within a bed; provision of an adequate description of bubble history is now one of the most pressing needs of fluidization engineering.

This model can, of course, be extended to noncatalytic reactors such as are of increasing interest in coal utilization.

ACKNOWLEDGMENT

This work has been assisted by grants from the Australian Research Grants Committee.

NOTATION

| | |
|-----------------|---|
| c_{avg} | = average reactant concentration in sample gas from probes |
| c_H | = reactant concentration in bed exit gas |
| c_0 | = reactant concentration in bed inlet gas |
| d_p | = particle diameter |
| D | = volume-equivalent bubble diameter |
| D_G | = gas phase diffusion coefficient |
| D_0 | = bubble diameter at distributor |
| f_w | = ratio of wake volume to bubble void volume |
| g | = gravitational acceleration |
| h | = vertical distance above distributor |
| H_{mf} | = height of bed at incipient fluidization |
| k | = first-order reaction rate constant, based on unit volume of dense phase |
| K_{BC} | = effective volumetric rate of gas exchange between bubble and cloud-wake per unit bubble volume |
| K_{BP} | = effective volumetric rate of gas exchange between bubble and particulate phase per unit bubble volume |
| K_{CP} | = effective volumetric rate of gas exchange between cloud-wake and particulate phase per unit bubble volume |
| m | = slope of linear relation between D and h |
| Q | = effective volumetric rate of gas exchange between bubble and dense phase |
| u_A | = bubble rise velocity in freely bubbling bed |
| U | = superficial velocity of fluidizing gas |
| U_{cr} | = critical value of U , above which backmixing occurs |
| U_{GB} | = superficial velocity of gas in bubble phase |
| U_{mf} | = superficial gas velocity at incipient fluidization |
| V | = volume of bubble void |
| ϵ_B | = volumetric fraction of bubbles in bed |
| ϵ_{mf} | = volumetric fraction of void spaces in bed at incipient fluidization |
| ρ_s | = particle density |

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A Model for Predicting Flow Regime Transitions in Horizontal and Near Horizontal Gas-Liquid Flow

Models are presented for determining flow regime transitions in two-phase gas-liquid flow. The mechanisms for transition are based on physical concepts and are fully predictive in that no flow regime transitions are used in their development. A generalized flow regime map based on this theory is presented.

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SCOPE

Predicting the flow regime for concurrent gas liquid flow in pipes has been a central unresolved problem in two-phase flow. The usual approach has been to collect data for flow rates and fluid properties and to visually observe the flow pattern through a transparent test section window. Then a search is undertaken for a way to map the data in a two-dimensional plot by locating transition boundaries between the regimes. This requires a decision to be made about the coordinates which are used. Because no theoretical basis for selection of coordinates has existed in the past, this approach represents a coordination of the data rather than a correlation and depends

strongly on the particular data being used to prepare the map. For this reason extension to other conditions of pipe size or inclination, fluid properties, and flow rates are of uncertain reliability.

This work has the objective of presenting a means for unambiguous analytical prediction of the transition between flow regimes based on physically realistic mechanisms for these transitions. The regimes considered are intermittent (slug and plug), stratified smooth, stratified wavy, dispersed bubble, and annular-annular dispersed liquid flow. The theory predicts the effect on transition boundaries of pipe size, fluid properties, and angle of inclination.

CONCLUSIONS AND SIGNIFICANCE

A theoretical model is developed which predicts the relationship between the following variables at which flow regime transitions take place: gas and liquid mass flow rates, properties of the fluids, pipe diameter, and angle of inclination to the horizontal. The mechanisms

for transition are based on physical concepts and are fully predictive in that no flow regime data are used in their development.

Five basic flow regimes are considered. When the theory is solved in dimensionless form the following