# GAS-LIQUID REACTORS. I. THE RESIDENCE TIME DISTRIBUTION IN MULTISTAGE BUBBLE REACTORS

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A general model is proposed for direct calculation of the residence time distribution in flooded multistage bubble reactors. The model is based on a sequence of imperfect mixers simulating the space between the plates. The most important parameters of the model are the magnitude of mixing between the stages (backmixing), the volume of dead region and the by-pass. The model has been tested on the water-air system in a 292 mm column and various geometrical arrangements of sieve plates with downcomers. The experiments indicate that in a majority of practical situations the flow of liquid in these reactors may be simulated adequately by a sequence of ideally mixed cells with backmixing.

A suitable, sufficiently general and simple model of the residence time distribution (RTD) in multistage bubble reactors with counter-current flow of phases has not been published to date. A concrete application of existing models is either limited or the models are unduly complex to be utilized directly as has been shown earlier<sup>1</sup>. The aim of this work was to propose a simple model of RTD based on real conditions in a multistage bubble reactor under counter-current flow of phases. The model should account for all effects causing deviations from ideal flow which can be reasonably expected in such equipment. The goal of the experimental part of the work was to gather enough data on RTD in a sieve-plate bubble column with downcomers under counter-current flow of phases. The experiments were carried out with the water-air system and a single diameter of the column. As variable parameters, whose effect on the hydrodynamics was examined, we took the geometrical characteristies of the plates (plate free area  $\varphi = 4 - 8\%$  and opening diameter  $d_0 = 1.6 - 5$  mm) and the flow rates of phases characterized by superficial velocity of gas ( $w_G = 0.008$  to 0.042 ms<sup>-1</sup>) and the volume of liquid supplied per unit area of column cross-section per unit time ( $W_{\rm L} = 2.5, 10^{-3} - 7.7, 10^{-3} \, {\rm m}^3 \, {\rm m}^{-2} \, {\rm s}^{-1}$ ). The ranges of  $w_{\rm G}$  and  $W_{\rm L}$ were taken so as to correspond to those commonly encountered in single-stage bubble reactors<sup>2</sup>

## THEORETICAL

In the formulation of the model we started from a qualitative concept of the flow of liquid in the given equipment, the plausibility of which had been tested by preliminary experiments. As a result we formulated a stage-wise model simulating the system by a sequence of imperfect mixers where each stage corresponds to a single stage of the real system. The stages are interconnected by the process stream and side streams. To account for non-ideal mixing in individual stages, the mixed region was combined with a dead region while a part of liquid may bypass the stage. The dead region is assumed to have a slow exchange of liquid with the ideally mixed part of the stage. Zero exchange flow rate then corresponds to a completely isolated dead region.

The model of a single stage of the sequence is thus that of Corrigan<sup>3</sup> proposed for single-stage stirred reactors. The bulk of the flow between the stages in the downcommers is modelled in our case by a sequence of ideal mixers with no backmixing. In addition, the model provides for the existence of side streams between the stages in both directions corresponding to the back flow and weeping through plate open-



ings. The existence of the side streams cannot be generally ruled out even in the top and bottom stages of the sequence but their magnitude here is apparently different from that within the column. The back flow from the top stage has a character of liquid carryover which is different than the back flow within the reactor. Liquid entrainment and weeping is assumed to exist only between the active (ideally mixed) regions of the stages. The mathematical model, the solution of which provides information about RTD of the liquid phase, thus represents a set of balance equation of a tracer inserted in a given instant into the process stream at a concentration Y. In the formulation

### FIG. 1

Scheme of RTD Model in Sieve-Plate Bubble Reactors of the balance equations we started from a flow diagram sketched in Fig. 1. The stages are numbered top-to-bottom. The overflows have the same number as the stage from which they draw liquid. The cells simulating the flow within the downcomer are also numbered top-to-bottom. (The first subscript indicates the number of the stage or its downcomer, the second one the number of the active or inactive region and the cell numbers in the downcomers.

The balance on the tracer for the first stage is

$$dY_{1,A}/d\Theta = K_2[a_2Y + EY_{2,A} + a_3Y_{1,D} - (a_2 + a_3 + E)Y_{1,A}], \qquad (1)$$

$$dY_{1,D}/d\Theta = K_3(a_3Y_{1,A} - a_3Y_{1,D}), \qquad (2)$$

$$Y_{1,C} = \left[ (1 - a_2) Y + (a_2 + E - G - e) Y_{1,A} \right] / (1 + E - G - e)$$
(3)

and for the n-th stage generally

$$dY_{n,A}/d\Theta = K_2 \{a_2(1 + E - G - e) Y_{n-1} + EY_{n+1,A} + GY_{n-1,A} + a_3Y_{n,D} - Y_{n,A}[G + E + a_3 + a_2(1 + E - G - e]\},$$
(4)

$$\mathrm{d}Y_{\mathrm{n},\mathrm{D}}/\mathrm{d}\Theta = K_3(a_3Y_{\mathrm{n},\mathrm{A}} - a_3Y_{\mathrm{n},\mathrm{D}})\,,\tag{5}$$

$$Y_{n,C} = (1 - a_2) Y_{n-1} + a_2 Y_{n,A}.$$
 (6)

For the N-th stage

$$dY_{N,A}/d\Theta = K_2 \{ a_2(1 + E - G - e) Y_{N-1} + GY_{N-1,A} + a_3 Y_{N,D} - Y_{N,A}[G + a_3 + a_2(1 + E - G - e)] \},$$
(7)

$$dY_{N,D}/d\Theta = K_3(a_3Y_{N,A} - a_3Y_{N,D}), \qquad (8)$$

$$Y_{N,C} = \{ [(1 + E - G - e) a_2 + G - g - E] Y_{N,A} + (1 - a_2) (1 + E - G - e) Y_{N-1} \} / (1 - e - g) .$$
(9)

The balance for a downcomer from the n-th stage

$$dY_{n,1}/d\Theta = K_4(1 + E - G - e)(Y_{n,C} - Y_{n,1}), \qquad (10)$$

$$dY_{n,n}/d\Theta = K_4(1 + E - G - e) (Y_{n,n-1} - Y_{n,n}), \qquad (11)$$

$$dY_{n,K}/d\Theta = K_4(1 + E - G - e) (Y_{n,K-1} - Y_{n,K}), \qquad (12)$$

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where  $Y_{n,K} = Y_n$  is the final concentration exiting from the downcomer and entering the (n + 1) th stage.

The constants  $K_i$  in Eqs (1) through (12) are defined as

$$K_2 = \bar{\iota} \dot{V}_{\rm L} / a_1 V_{\rm Li} \,, \tag{13}$$

$$K_3 = i \dot{V}_{\rm L} / (1 - a_1) V_{\rm Li} , \qquad (14)$$

$$K_4 = \bar{\iota} \dot{V}_L / K_1$$
, (15)

$$K_1 = V_P / K$$
. (16)

The mean residence time of the liquid phase is related to the whole reactor volume  $V_{\rm L}$ 

$$V_{\rm L} = \sum_{i=1}^{\rm N} V_{\rm Li} + (N-1) V_{\rm P}$$
(17)

or

$$V_{\rm L} = V_{\rm r} (1 - \varepsilon_{\rm G}) \,. \tag{18}$$

In the formulation of Eqs (1)-(12) the internal entrainment and weeping were assumed equal throughout the reactor. This assumption need not be generally satisfied and corresponding values E and G in such a case would have to be replaced by  $E_{n,n+1}$ ,  $G_{n-1,n}$  for each pair of plates.

A full mathematical statement represents (3N + K(N - 1)) equations. By solving this set for the feed signal Y given by the Dirac function

$$Y = 0$$
,  $\Theta \ge 0$ ;  $Y = \infty$ ,  $\Theta = 0$ ;  $\int_{0}^{\infty} Y d\Theta = 1$  (19)

one obtains a function  $Y_N = Y_N(\Theta)$  representing a response of the model to a pulse. This response corresponds to the distribution function of the residence time at the exit from the system (E-curve) carrying information about the character of the flow within the system<sup>4</sup>. Plots of  $Y_N = Y_N(\Theta)$  will therefore be referred to in the following as E-curves. The proposed model contains 9 parameters  $(a_1, a_2, a_3, E, e, G, g, N, K)$  all of them being physically meaningful. The parameters N and K characterize the arrangement of the system and the rest the effect of various types of flows and mixing regions on RTD which plausibly may exist. In the application of the model for determing the maximum conversion or optimum operating conditions for a given reaction in a real reactor the large number of the parameters would not be convenient both for the necessity of having a large number of correlations available and/or for the complexity of the calculation of RTD itself. In such a case though the parameters N and K are fixed by the arrangement of the real reactor. In addition, all the types of flows and mixing regions that had been implemented in the model may not exist, or may be neglected and the number of parameters may thus be further reduced.

Generally, one must expect that in large diameter reactors the gradient in horizontal level is not negligible and thus the concept of the behaviour of a stage as a single ideal mixer with dead region and bypass may not be adequate to the real conditions. In such a case one can change to a cascade of ideally mixed cells interconnected by the process stream and backmixing in the horizontal direction<sup>1</sup>. A part of the volume of each cell is occupied by the dead region and the cell may have a bypass. A sketch of the model with these additional features is shown in Fig. 2 and its mathematical formulation can be made on the basis of Eqs (1) - (12).



FIG. 2 Scheme of the Extended RTD Model

The full mathematical statement of the problem represents a set of N(2P + 1) + K(N - 1) equations to be solved together with the conditions in Eq. (19). The solution is a function  $Y_N = Y_N(\Theta)$  for a given set of parameters.

In contrast to the first model proposed in this paper we have two more parameters: the number of cells within the stage (P) and the coefficient of backmixing between the cells (R). The values of these parameters, which make the problem considerably more complex, are apparently very difficult to determine by independent experiments and as follows from the analysis<sup>1</sup> their certain combinations in relatively wide intervals provide almost equal RTD. This is, of course, at odds with the requirements put on the model of RTD formulated earlier.

There are no data available thus far which would suggest the necessity to change to this model, although from the viewpoint of approximating to the piston flow the regime described by this model would be desirable. It seems that such situation would occur if we used suitable baffles occupying the space between the plates to direct the flow within the stage. In such a case the number of cells and the magnitude of backmixing could be estimated and the drawback associated with the application of this model removed.

Preliminary experiments, carried out with baffles in the space between the plates of a bubble reactor which forced the liquid to follows a longer path from the inlet of the plate toward the wall and back before reaching the downcomer, seem to confirm this idea. The space between the vertical baffles on the plate would correspond roughly to an ideally mixed cell.

## EXPERIMENTAL

The basic set of experiments corresponds to a factorial experiment with doubled duplication designed to examine the effect on RTD of liquid of four quantities ( $w_G$ ,  $W_L$ ,  $d_0$ ,  $\varphi$ ) on three, or two levels.

Apparatus. The experiments were carried out in a sieve-plate column with downcomers. The sketch of the set-up is shown in Fig. 3. The column consisted of three stages formed by glass cylinders 1 mounted by flanges 2. The individual stages were separated by plates mounted in an nuli 3 held by the flanges. The downcomers 4 were arranged so as to force the liquid to flow perpendicularly to the flow of gas. The inner diameter of the glass cylinders was 0-292 m; the spacing of the plates 0-464 m. The height of liquid in the top stage vented in the atmosphere was held at a constant level by an overflow vessel 5. A 0·35 m high stainless-steel cylindrical chamber 6 of the diameter equaling that of the glass cylinders supported the bottom plate. The bottom of the chamber was tilted toward a tap 7 used to discharge the collected liquid into a measuring cylinder 8. The arrangement of the flows of the phases was counter-current, the gas phase being air brought via pressure regulator 9 and oil filter 10. The air loop was further equipped with a humidifer 11 and a rotameter 12. The selected flow rate was held constant by a pneumatic valve 13 controlled by a regulator 14. The regulated quantity was the pressure forg across an orifice 15 measured on a manometer 16 providing a signal for the regulator. The air was fed sideways into the chamber ber below a gas distributing plate 17 mounted about 0·1 m below the bottom plate. The free

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area of the distributing plate was 1%; the 1.6 mm openings were arranged in a square pitch. The gas rotameter was calibrated by means of a gasometer. The experimental values of  $w_{G}$  are arithmetic averages of superficial velocity near the head and the bottom of the column. Tap water was used as liquid supplied into a storage tank 18 and driven by a pump19 *via* glass pipe into a rotameter 20 on the top of the column. The rotameter was calibrated by weighing the liquid drained after a sufficiently long interval. The calibration was checked repeatedly during experiments. The accuracy of setting the flow rate was within 3%. The liquid entered the top stage through two tubes of the diameter equalling that of the downcomers. The bottom stage was drained by a pubcolorimetric cell 22 and an overflow vessel to be either returned into the storage tank or discharged. There were 2 mm tubes with cocks in the flanges of all three stages enabling the tracer solutions to be introduced below the mouth of the downcomers or below the plates in individual stages.

*Plates.* The plates used were sieve plates with downcomers which did not protrude above the level of the plate. The function of the downcomers thus were not that of overflow weirs. There were two 0.04 m in diameter 0.42 m long downcomers next to each other on each plate. The length of the downcomers in some experiments was 0.1 m. Both the plates and the downcomers were made of brass; the thickness of the plates was 0.002 m. The openings of the plates were circular with unchmafered edges. The openings were arranged in a square pitch covering only the central area of the plate<sup>5</sup>. On one side of the plate opposite to the downcomers there was an identical area without openings facing the openings of the downcomers from the above stage.

Measurement of the residence time distribution. The data on RTD in the column were obtained in the form of responses to a pulse input (responses referred to as C-curves by Levenspiel<sup>4</sup>). In accord with the term for the curves obtained by solving the mathematical model of RTD these curves, indicating the distribution of the residence time, are termed E-curves. A colorimetric method with methyl blue as a tracer was used to obtain the response curves. This tracer, which is commonly used for such purposes<sup>6,7</sup>, was chosen as most suitable by test from a number of chemicals (cosin, fluorresceine, sulphomalachite green). The impulse was simulated by injecting 10 ml of methylene blue solution ( $c_0 = 10$  g/l). The tracer solution was injected by a syringe into the







stream entering the top stage (below the mouth of the downcomers). The effluent concentration of methylen blue was determined by measuring optical transmissivity. The liquid passed through the cell between a light source and a photoelectric cell. A constant value of the current was checked by miliampermeter connected into the circuit. The photoelectric current was recorded on a Kipp-Zonen recorder and the curves were transformed computationally to give the true concentration response. The transformation was necessitated by non-linearity of the recorder. The obtained responses were further rendered dimensionless on a computer to a form corresponding to a unit pulse input.

Accuracy. Under our experimental conditions (liquid hold-up in the column was 801) the injected volume of the tracer did not affected the regime within the column. In order that the error due to imperfect impulse<sup>8</sup> may be neglected it is usually recommended that  $\Delta t/\bar{t} < 0.05$ . In our case it stipulates that  $\overline{i} > 160$  s, which corresponds to  $\Delta i < 8$  s. This condition was well met in all experiments. Injection of the tracer solution was carried out so as to ensure a uniform dispersion of the tracer in the incoming stream of liquid. The colorimetric method in practice measures average concentrations of the tracer in the outgoing stream and thus eliminates possible errors due to localized measurement. Preliminary experiments revealed no time lag of the recorder behind the concentration changes. The effect of the position of the probe being 0.40 m from the column exit was compensated in the computer program for theoretical E-curves by simulating the exit tube (similarly as the downcomers) by a sequence of three ideal mixers without backmixing. The reproducibility of the procedure intended for obtaining the E-curves was tested by duplicating each experiment of the basic set. The relative deviations were computed of the characteristic values ( $Y_{\theta=1}, \Theta_{Y, max}$ ) obtained from experimental E-curves under identical conditions ( $W_{\rm L}$ ,  $w_{\rm G}$ ,  $d_0$ ,  $\varphi$ ) from the average of the duplicated experiments. The average relative deviations of all pairs amounted to  $\overline{\delta}_{\Theta Y, max} = 2 \cdot 1\%$ ,  $\overline{\delta}_{Y\Theta_{=1}} = 0.6\%$  while the maximum relative deviations equaled  $(\delta_{\Theta Y, \max})_{\max} = 9.5\%, (\delta_{Y\Theta_{-1}})_{\max} = 1.9\%$ 

### **RESULTS AND DISCUSSION**

A quantitative expression of the effect of the model parameters on RTD may be obtained by solving the model for various values of individual parameters. Owing to the laboriousness of the solution the analysis could not be made for all parameters of the model. The selection of the parameters to be examined was made with the following aspects in mind: 1) Expected importance or degree of effect on RTD, 2) probability of existence of the effect in real equipment, or the necessity to take this effect into consideration, 3) the existence of the effect in the experimental equipment. With regard to the possibility of comparing the results of the analysis with the experimental data the third aspect was virtually decisive, although in view of the experimental set-up being a fairly representative specimen of the bubble reactor it did not pose any serious restriction of the choice of parameters. The selected parameters were the effect of backmixing, the bypass, the volume of the dead region and the rate of exchange between the dead region and the ideally mixed part of the stage. This choice thus encompasses all three parameters describing the mixing in individual stages and the coefficient of backmixing characterizing the degree of mixing between individual stages. For the values of other parameters appearing in the model we substituted those corresponding to the conditions in the reactor. It may be as-

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sumed that under the flow rates of phases common in bubble reactors no appreciable entrainment from the top stage occurs. Our measurements<sup>5</sup> have confirmed this assumption and further showed that in a wide range of operating conditions the weeping within the column may also be neglected. The values of the parameters e,g and Gwere thus put equal zero. In accord with the number of the stages in the experimental column we put N = 3 and the number of cells simulating the flow in the downcomers was taken K = 3. Generally, various values of K should be taken so as to correspond to various conditions in a real reactor. For the Reynolds number, Re, characterizing the flow within the downcomers under the given conditions one can read  $D/v_{LP}L$  off the graph<sup>4,9</sup> and calculate the appropriate number of cells from

$$\frac{1}{K} = 2 \frac{D}{v_{LF}L} - 2 \left(\frac{D}{v_{LF}L}\right)^2 \left[1 - \exp\left(-\frac{v_{LF}L}{D}\right)\right].$$
(20)

This equation enables transition from a dispersion model to a sequence of ideal mixers. The values of K substituted into the relations were computed for the conditions under which the experimental E agrees with that used in the computation. With increasing number of cells, however, the computer costs increase while the effect of the number of cells on the response is growing small owing to a small down-comer-to-total volume ratio. Comparative computations showed three cells to approximate well even cases where the correct number of cells was 10 or 11. Thus the theoretical results for K = 3 may be compared with the experimental data. After



substituting for g, G, e, N and K the set of equations representing the mathematical model was solved for various E,  $a_1$ ,  $a_2$ ,  $a_3$  ranging:  $E \in \langle 0; 6 \rangle$ ,  $a_1 \in \langle 0; 5; 1 \rangle$ ,  $a_2 \in \in \langle 0; 5; 1 \rangle$ ,  $a_3 \in \langle 0; 0; 5 \rangle$ . The result of the solution was a set of E-curves corresponding to RTD as a function of the selected parameters. For a quantitative expression of these relations we had to choose convenient characteristics of E-curves which would respond selectively to the changes of E,  $a_1$ ,  $a_2$ ,  $a_3$ . There are several alternatives existing in the literature<sup>1,8,10</sup>. It is apparent that taking e.g. the variance of the E-curves and hence RTD in dependence on the examined parameters we took the abscissa of the maximum of the curve ( $\Theta_{Y,max}$ ) and the ordinate corresponding  $\Theta = 1$  ( $Y_{\Theta=1}$ ). The values of these quantities obtained as a solution of the mathematical model of RTD were plotted for various values of the parameters ( $a_1$ ,  $a_2$ ,  $a_3$ , E) to obtain a graph  $Y_{\Theta=1}$  versus  $\Theta_{Y,max}$  illustrating their effect on RTD. A typical plot is shown in Figs 5a, b.

Fig. 4 or 5 enable a qualitative and quantitative estimate of liquid mixing to be made on the basis of experimental RTD obtained in the form of E-curve. For a pair of values  $Y_{\Theta=1} - \Theta_{Y,max}$ , characterizing a given experimental curve, one can find from the graphs which of the mixing regions and flow types, which had been incorporated into the model, exist in the given equipment and, eventually, determine



FIG. 5

 $Y_{\Theta=1}$  and  $\Theta_{Y_{e},\max}$  as Functions of the Model Parameters

 $\begin{array}{c} a \ 1 \ a_1 = 1, \ a_2 = 1; \ 2 \ a_1 = 1, \ a_2 = 0.95; \ 3 \ a_1 = 1, \ a_2 = 0.9; \ 4 \ a_1 = 1, \ a_2 = 0.75; \ 5 \\ a_1 = 1, \ a_2 = 0.5; \ 6 \ a_1 = 0.9, \ a_2 = 1; \ 7 \ a_1 = 0.75, \ a_2 = 1; \ 8 \ a_1 = 0.5, \ a_2 = 1; \ 9 \ a_1 = 0.75; \ a_2 = 0.75; \ 5 \ 1 \ a_1 = 0.5, \ a_2 = 0.75; \ 5 \ 1 \ a_1 = 0.75; \ a_2 = 0.75; \ 1 \ a_1 = 0.75; \ a_2 = 0.75; \ 1 \ a_1 = 0.75; \ a_2 = 0.75; \ a_1 = 1, \ a_2 = 0.75; \ a_2 = 1; \ a_1 = 1; \ a_2 = 0.75; \ a_2 = 1; \ a_1 = 1; \ a_2 = 0.75; \ a_1 = 1, \ a_2 = 0.75; \ a_2 = 1; \ a_1 = 0.75; \ a_2 = 1; \ a_2 = 0.75; \ a_2 = 1; \ a_3 = 0. \end{array}$ 

their magnitude. From Fig. 4 it is apparent though that such assessment is not unambiguous because the functions  $Y_{\theta=1} - \Theta_{Y,max}$  corresponding to various combinations of the parameters mutually intersect and hence a single point  $(Y_{\theta=1}; \Theta_{Y,max})$  may corespond to several parameter combinations. This difficulty cannot be avoided by any alternative choice of the characteristics of the E-curves. Thus for reliable results additional information is needed about the conditions within the reactor, *i.e.* one more real value of some of the model parameters. For an experimentally determined parameter and a pair of  $Y_{\theta=1} - \Theta_{Y,max}$  one can then evaluate unambiguously the remaining parameters and thus determine the character of mixing within the equipment. The other alternative is to compare the E-curve computed for the parameters read off Fig. 4 with the experimental curve. The method requires fitting of the whole curve and repeating the whole procedure for a new set of parameters to improve the fit. This method would be suitable only in cases when for some reason no other parameter can be measured.

The coefficient of backmixing between the stages, E, seems most suitable of all parameters whose effect on RTD have been examined for separate determination since the methods for its evaluation independently on RTD are numerous, *e.g.* cit.<sup>8,11</sup>.

Moreover, under the conditions of this work (*i.e.* a sieve-plate column with downcomers and counter-current flow of phases) it is the coefficient of backmixing that affects RTD of liquid most markedly of all parameters.

A total of 160 experimental E-curves were processed for various values of  $W_{\rm L}$ ,



FIG. 6

 $\Theta_{Y,max}$  as a Function of the Model Parameters  $a \varphi = 4\%$ ,  $b \varphi = 8\%$ 

Theoretical curves: 1  $a_1 = a_2 = 1$ ,  $a_3 = 0$ ; 2  $a_1 = 0.95$ ,  $a_2 = 1$ ,  $a_3 = 0$ . Experimental points  $-d_0$ :  $\bullet 1.6$ ,  $\oplus 3.0$ ,  $\odot 5.0$ .

 $w_{G}$ ,  $d_{0}$  and  $\varphi$ . Figs 6a,b show a comparison of the experimental  $Y_{\Theta=1} - \Theta_{Y, \max}$  values with those obtained by solving the mathematical model of RTD for various combinations of  $a_1, a_2, a_3, E$ . From comparison it is seen that RTD of liquid approaches for all values of the geometrical parameters  $d_0$  and  $\varphi$  that of a sequence of three ideal mixers with backmixing between the stages. The backmixing decreases with increasing  $W_{\rm L}$  and decreasing values of  $w_{\rm G}$ ,  $d_0$  and  $\varphi$ . The existence of bypasses may be neglected in the whole range of the examined parameters. Under high liquid velocities  $(W_{\rm I} = 7.5 . 10^{-3} \text{ m}^3/\text{m}^2 \text{ s})$  and low gas velocities  $(w_{\rm G} = 0.008 \text{ or } 0.025 \text{ s})$ m/s) the data point at the existence of the dead region isolated from the active volume of the column (with no exchange of mass between the dead region and the bulk) regardless of  $d_0$  and  $\varphi$ . The existence of an entirely isolated dead region is in accord with the visual observation of the flow of phases in the column. With increasing  $W_{\rm I}$  and decreasing  $w_{\rm C}$  the ascending stream of gas narrows and the number of the disturbances decreases that bring about random whirling of liquid breaking basic pattern of liquid circulation and contributing to mixing. Simultaneously we have observed also a decrease of the back flow with increasing  $W_{\rm L}$  and decreasing  $w_{\rm G}$ . Thus it may be assumed that under these conditions the intensity of liquid phase mixing in individual stages slackens and certain regions bypassed by bubbles outside the circulating flow of liquid may appear. For the magnitude of the dead region we read off the graph in all cases  $a_1 = 0.95$ . In order to avoid interpolation, the values of the coefficient of the back flow corresponding to the experimental E-curves were read off for individual  $\Theta_{Y,max}$  from the theoretical  $\Theta_{Y,max}(E)$  relation for  $a_1 = 1.0$ (or  $a_1 = 0.95$ ),  $a_2 = 1$  and  $a_3 = 0$ . Table I is a comparison of the values of E obtained as just described with the experimental ones measured under identical conditions<sup>5</sup> ( $W_{\rm L}, w_{\rm G}, d_0, \varphi$ ) as the corresponding E-curves. The comparison evidences a good agreement of the coefficients (the maximum deviation is about 10%) which



Fig. 7

A Comparison of Some Experimental and Computed E-curves

Theoretical curves: 1 E = 3·3,  $a_1 = 1$ ,  $a_2 = 1$ ,  $a_3 = 0$ ; 2 E = 0·57,  $a_1 = 1$ ,  $a_2 = 1$ ,  $a_3 = 0$ ; 3 E = 0·12,  $a_1 = 0.95$ ,  $a_2 = 1$ ,  $a_3 =$ = 0. Experimentals points: •  $\varphi = 8\%$ ,  $d_0 =$ = 3 mm,  $W_L = 2 \cdot 5 \cdot 10^{-3} \text{ m}^3/\text{m}^2 \text{ s}$ ;  $w_G =$ = 0·042 m/s; •  $\varphi = 4\%$ ,  $d_0 = 3 \text{ mm}$ ,  $W_L =$ = 5·0 · 10<sup>-3</sup> m<sup>3</sup>/m<sup>2</sup> s,  $w_G = 0.008 \text{ m/s}$ ;  $\oplus$  $\varphi = 4\%$ ,  $d_0 = 3 \text{ mm}$ ,  $W_L = 7 \cdot 5 \cdot 10^{-3} \text{ m}^3/\text{m}^2$ . further supports the model serving to obtain E and  $a_1$  from the graphs (a sequence of ideal mixers with the dead region and backmixing between individual stages). This finding has been confirmed also by comparing the experimental *E*-curves with the theoretical ones computed for corresponding E and  $a_1$ . Typical pairs are compared in Fig. 7.

From the analysis of the dependences of  $Y_{\theta=1}$  and  $\Theta_{Y,max}$  on individual variables<sup>5</sup> one can estimate qualitatively the effect of the examined quantities on the mixing of liquid in the column. Since in the examined range of v. ri bles the existence of the by-pass and in practice also the existence of the dead region may be neglected RTD of liquid is sufficiently characterized by  $\Theta_{Y,max}$  and the quantitative effect of individual quantities is sufficiently determined by their effect on  $\Theta_{Y,max}$ . By processing the experimental data as a function of  $W_L$ ,  $w_G$  and  $d_0$  for individual  $\varphi$  the following empirical relation was obtained

$$\Theta_{Y,\max} = L_1 w_G + L_2 \log W_L + L_3 \log d_0 + L_4, \qquad (21)$$

where

A comparison of the experimental  $\Theta_{Y,max}$  values with those calculated from Eq. (21) shows a good agreement. The relative deviation amounts to 1.4%. Thus Eq. (21) can be used to estimate quantitatively the effect of  $W_L$ ,  $w_G$ ,  $d_0$  and  $\varphi$  on RTD of liquid in the investigated range.

In our case, where there were no bypasses and the dead regions existed only near the limits of the investigated range of flow rates in the extent corresponding to only negligible effect of RTD on reaction conversion, the problem of getting suitable expressions for the model parameters thus reduces to that of finding an equation for the coefficient of backmixing. On combining Eq. (21) with an analytical expression of the theoretical function  $\Theta_{Y,max} = \Theta_{Y,max}(E)$  for the case of no bypass and no dead region

$$\Theta_{Y,max} = -0.404 \log (E + 0.314) + 0.482 \text{ for } E \in \langle 0; 4 \rangle$$
(22)

we get for the coefficient of backmixing

$$E = M_1 M_2^{w_0} W_1^{M_3} d_0^{M_4} - 0.314 , \qquad (23)$$

where

$$M_1 = 7.478 \cdot 10^{-4}$$
,  $M_2 = 3.35 \cdot 10^8$ ,  $M_3 = -1.230$ ,  $M_4 = 0.325$ ,  
for 4%;

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TABLE I

Measured and Calculated Coefficients of Backmixing

φ	$d_0$	$W_{\rm L}$ . 10 <sup>3</sup>	WG.	E		5. M	$\Delta E$ 100	
%	mm	$m^3/m^2s$	m/s	exp	graph	$a_1$	$E_{exp}$ . 100	
4	1.6	2.5	0.008	1.327	1.275	1	3.9	
		2.5	0.025	2.100	1.950	1	7.1	
		2.5	0.042	3.063	2.720	1	11.1	
		5.0	0.008	0.435	0.420	1	3.4	
		5.0	0.025	0.673	0.730	1	10.0	
		5.0	0.042	1.082	1.090	1	0.7	
		7.5	0.008	0.066	0.060	0.95	9.1	
		7.5	0.025	0.278	0.290	0.95	4.3	
		7.5	0.042	0.525	0.595	1	13.3	
4	3	2.5	0.008	1.847	1.700	1	8.0	
		2.5	0.022	2.515	2.540	1	1.0	
		2.5	0.042	3.375	3.260	1	3.4	
		5.0	0.008	0.510	0.570	1	11.8	
		5.0	0.025	0.868	0.880	1	1.4	
		5.0	0.042	1.421	1.360	1	3.3	
		7.5	0.008	0.121	0.120	0.95	0.8	
		7.5	0.022	0.362	0.345	0.95	4.7	
		7.5	0.042	0.706	0.700	1	0.8	
4	5	2.5	0.008	1.055	1.980	1	1.3	
4	5	2.5	0.025	2.802	2.120	1	8.2	
		2.5	0.042	3.854	3.720	1	3.7	
		5.0	0.008	0.643	0.620	1	5-9	
		5.0	0.025	1,070	1.060	1	1.8	
		5.0	0.042	1.615	1.480	1	9.4	
		7.5	0.008	0.220	0.245	0.05	11.4	
		7.5	0.025	0.472	0.505	0.95	7.0	
		7.5	0.042	0.788	0.840	0.95	6.6	
		, 5	0012	0 / 00	0 0 10	0 75	00	
8	1.6	2.5	0.008	1.796	1.800	1	0.2	
		2.5	0.025	2.381	2.220	1	6.8	
		2.5	0.042	3.014	2.760	1	8.4	
		5.0	0.008	0.675	0.724	1	7.3	
		5.0	0.025	0.949	0.880	1	7.3	
		5.0	0.042	1.124	1.100	1	2.1	
		7.5	0.008	0.292	0.300	0.95	2.7	
		7.5	0.025	0.459	0.420	0.95	8.5	
		7.5	0.042	0.737	0.670	1	9.1	

TABLE I

(Continued)

%	<i>d</i> <sub>0</sub> mm	$W_{\rm L} \cdot 10^3$ m <sup>3</sup> /m <sup>2</sup> s	₩ <b>G</b> m/s	Ε		$a_1$	$\frac{\Delta E}{E_{\rm exp}}$ . 100
8				2.279	2.100	1	7.9
		2.5	0.025	2.987	2.720	1	8.9
		2.5	0.042	3-525	3.300	1	6.4
		5.0	0.008	0.820	0.880	1	3.5
		5.0	0.025	1.109	1.055	1	4.9
		5.0	0.042	1.316	1.350	1	2.6
		7.5	0.008	0.403	0.400	0.95	0.7
		7.5	0.025	0.533	0.510	0.95	4.3
		7.5	0.042	0.816	0.790	1	3.2
8	5	2.5	0.008	2.553	2.450	1	4.0
		2.5	0.022	3.353	3.130	1	6.7
		2.5	0.042	3.867	3.720	1	3.8
		5.0	0.008	0.985	1.020	1	3.6
		5.0	0.025	1.217	1.280	1	5.2
		5.0	0.042	1.556	1.475	1	5.2
		7.5	0.008	0.493	0.490	0.95	0.6
		7.5	0.025	0.637	0.640	0.95	0.5
		7.5	0.042	0.912	0.910	1	0.5

and

The average relative deviation from experimental  $E^5$  is 6.7%. Owing to the complexity of Eq. (23) it may be more convenient to determine E from the graph  $\Theta_{Y,max} - E$  (e.g. in Fig. 5) for  $\Theta_{Y,max}$  computed from Eq. (21).

The proposed model of the distribution of the residence time of liquid may be used to calculate the limiting value of the reaction conversion. In this case, however, one has to regard the liquid as a micro-fluid and the applicability of the Danckwerts integral is limited<sup>4</sup>. On the other hand, the experimental verification of the model has shown that in flooded multi-stage columns less than 300 mm in dimaeter and the height of the stage ranging below 460 mm the space within the stage may be regarded as practically ideally mixed. Under the flow rates of gas and liquid common in practice the calculation of the resulting reaction conversion on the exit of the reactor may then

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be carried out by a simple stage-to-stage balance taking into consideration the backmixing between the stages, *e.g.* from Eqs (22) and (23). Since the data from this region are lacking these equations may be used for an arbitrary system as a first approximation provided that they do not depend strongly on physicochemical properties of liquid.

## LIST OF SYMBOLS

- a1 ratio of ideally mixed to total volume of the stage
- a2 ratio of flow rate in ideally mixed part to total flow rate
- a3 ratio of flow rate between ideally mixed part and dead region to total flow rate
- c concentration  $(M/L^3)$
- D axial dispersion coefficient  $(L^2/T)$
- d<sub>0</sub> plate opening diameter (L)
- $d_{\rm p}$  downcomer diameter (L)
- $\vec{E}$  ratio of back flow between two adjacent stages to total flow rate
- E residence time distribution function (E-curves)
- G ratio of weeping between two adjacent stages (internal weeping) to total flow rate
- g ratio of weeping from bottom stage (external weeping) to total flow rate
- K number of ideally mixed cells
- $K_1, K_2, K_3, K_4$  coefficients defined by Eqs (13)-(16)
- L length of downcomer (L)
- N number of stages or number of cells in model sequence
- P parameter of RTD model
- R parameter of RTD model
- t time (T)
- $\Delta t$  duration of tracer injection (T)
- V volume (L<sup>3</sup>)
- $\dot{V}$  volume flow rate (L<sup>3</sup>/T)
- $V_{\rm p}$  volume of downcomers in a one stage (L<sup>3</sup>)
- v real velocity of phases (L/T)
- $v_{LP}$  velocity of liquid in downcomer (L/T)
- w superficial velocity of phases (L/T)
- $W_{\rm L}$  superficial velocity of liquid  $(L^3/{\rm L}^2{\rm T})$
- Y dimensionless concentration
- $\delta$  relative deviation
- εG porosity of bubble layer
- $\Theta$  dimensionless time
- $\varphi$  relative free area of plate

### Subscripts

- |
- A active part of stage
- D dead region
- G gas
- L liquid
- i i-th quantity

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