MASS TRANSFER IN BUBBLE-TYPE REACTORS WITHOUT MECHANICAL MIXING

F.KAŠTÁNEK, J.KRATOCHVÍL and M.RYLEK

Institute of Chemical Process Fundamentals,
Czechoslovak Academy of Sciences, 165 02 Prague 6 - Suchdol

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Experimental volumetric mass transfer coefficients k_La in bubble type reactors are given. Reactor sizes were 0·15, 0·3 and 1·0 m, bed heights up to 1·2 m. Superficial gas velocities were up to 0·03 m/s. Coefficients k_La were measured by physical absorption of oxygen from air into water. Oxygen was removed from water in advance by addition of sodium sulphite solution and cobalt salt as catalyst.

Under the experimental conditions studied the reactor size was found to have no effect on k_La. Except of the single point gas inlet into the liquid, the type of distributor situated at the bottom of the bed has no effect on k_La.

New experimental data, obtained in reactors of sizes close to industrial dimensions, should give an answer to the basic question as to what is the effect of scaling up of bubble-type reactors on mass transfer. This answer should be also useful for extrapolation of laboratory data. Measurements of accurate data in larger reactors are more time consuming and costly which might be one of the reasons why the study on scaling up has not been paid appropriate attention yet.

Here, an attempt has been made to fill, at least partially, this gap by experiments on mass transfer in bubbled beds with sizes up to ID 1 m.

EXPERIMENTAL

Reactors: Cylindrical columns with the inside diameters $D_K = 150$ and 300 mm and the rectangular section 1 × 1 m were used. Gas distributors were *1*. Perforated plates with $d = 1·6$ mm, $\varphi = 0·005$ for all used column sizes. *2*. Spider type distributor (for section 1 × 1 m). On the horizontal distributing pipe of ID 80 mm situated in parallel with the side of the section were perpendicularly welded perforated pipes with ID 25 mm. The distributor was situated at the bottom of the section with the gas inlet into the distributing pipe. Distances of individual pipes were: reactor wall — 100, 200, 200. 200, 100 mm — reactor wall. Circular holes 4 mm in diameter were drilled in the pipes, situated according to Fig. 1, on their lower side and were pointed toward the bottom of the section. The total number of holes was 330, free plate area (ratio of all holes in the pipes to the cross-sectional area of the reactor) was $\varphi = 0·004$. Direction of the outlet of gas from the distributor was toward the bottom of the section and its distance from the bottom was 50 mm. *3*. Eccentric gas inlet with one hole 50 mm in dia in the side-wall of the reactor situated 50 mm from the bottom and 200 mm from the corner of the reactor (for the reactor 1 × 1 m^2). Gas flow rate was measured by callibrated rotameters and callibrated orifices.

Measurement of volume mass transfer coefficient $k_L a$: In the continuous arrangement with columns 150 and 300 mm; batchwise in unsteady state with the gas flow in the reactor 1 000 mm. Column 150 mm: liquid flow rate 8·6 l/min, bed height 600 mm. Column 300 mm: liquid flow rate 9·4 l/min, bed height 600 mm. Column 1 000 mm: bed heights 600 to 1 200 mm. The mass transfer coefficient $k_L a$ is related to the unit volume of liquid on the plate. Liquid was introduced into the columns 150 and 300 mm by a pipe provided with the bubble-cap to reverse the direction of the liquid flow at the distance of 45 mm from the plate. Liquid was discharged through the downcomer that was fixing the required liquid height.

The samples were taken for analysis at two positions in the wall of the reactor, namely, 65 and 375 mm from the bottom, after having reached the steady operation of the reactor at the given gas flow rate. From each position were taken at least 4 samples at 3 min intervals. The analysis of oxygen content has proved the practically ideal liquid mixing. Points in Fig. 2 are representing the average of at least 8 experiments. The liquid was tap water which was deprived of oxygen by addition of about 0·7 to 0·8 g/l of anhydrous $Na_2SO_3 + CoSO_4$ (in total, $5 \cdot 10^{-5}$ mol/l) as a catalyst. The content of oxygen was measured until the final inlet concentration of about 0·15 mg/l of oxygen was reached. Simultaneously with the sampling the inlet concentration of oxygen was checked.

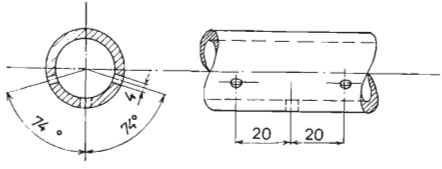

FIG. 1
Horizontal Pipe of the Distributor

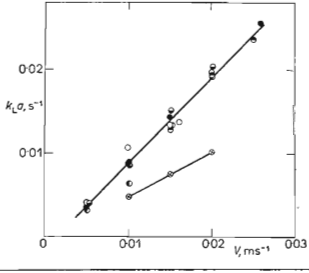

FIG. 2
Volume Mass Transfer Coefficient $k_L a$ in Dependence on the Column Diameter and Type of Gas Distributor

● Column 1 000 mm, perforated plate, d 1·6 mm, φ 0·005 h 1 200 mm. ○ Column 1 000 mm, perforated plate, d 1·6 mm, φ 0·005, h 600 mm. ◐ Column 1 000 mm, spider type distributor, h 1 200 mm. ◓ Column 150 mm, perforated plate, d 1·6 mm, φ 0·005, h 600. ◒ Column 300 mm, perforated plate, d 1·6 mm, φ 0·005 ,h 600 mm. ⊗ Column 1 000 mm, eccentrically situated hole for gas inlet 50, h 1 200 mm.

In the batchwise procedure, the tested amount of 150—300 g/1 000 l of Na_2SO_3 (in proportion to the bed height and gas flow) + $CoSO_4$ ($5 \cdot 10^{-5}$ mol/l) in aqueous solution were introduced (in excess) into the bubbled bed of tap water and samples were being taken at 15 s intervals under continuous bubbling.

The sampling proceeded through the point of zero oxygen concentration and further until the liquid was saturated. From the curve "oxygen content–time" k_La values were calculated by the statistical method of trial and error and were substituted into the theoretical balance equation. The corresponding theoretical concentrations were compared to those experimental so that the sum of squared standard deviations was minimal. The calculation of k_La assumed ideal liquid mixing in all reactor size.

Determination of oxygen: As the most suitable was considered the classical[1] method: samples were taken into flasks with slant stoppers which enabled immediate removal of eventual bubbles present. After addition of solution of $MnSO_4$ and soln. KOH, KJ and Na_3N the sedimented precipitates were dissolved by diluted sulphuric acid and were titrated by sodium thiosulphate in the presence of starch solution. The oxygen content was calculated from stoichiometry. Equilibrium solubilities of oxygen were related to the temperature of individual experiments and were in the range from 16 to 19°C.

Measurement of porosity: Porosity was determined manometrically by pressure measurements at two levels (at the vessel bottom and at the upper foam surface).

Measurement of the cross-sectional area of the upward liquid flow and of its velocity: The cross-sectional area available for the circulating liquid flow was measured visually by reading off the width of the circulating stream on the scale in the middle of the bed at the moment the circulation stream became steady. Each measurement was repeated several times and the average was calculated. In circular columns the area was calculated from the width of the circular segment. In square column the linear dimension corresponds to the area for flow. Measurements were performed at two bed heights of 600 and 1 200 mm with the water–air mixture. The linear velocity of the circulating stream in the limited section along the bed height was measured by use of tracer particles made of polyethylene with their specific density fixed experimentally (by addition of water) so that they corresponded to that of water. The size of tracer particles was chosen so as not to affect the character of streaming ($D_{T\max}/D_C < 0{\cdot}15$).

RESULTS

In the range of superficial gas velocities from 0 to 0·03 ms^{-1} and in the range of measured geometrical parameters of the bed the so-called single-loop circulation as described earlier[2] has developed in all cases. Development of this circulation in the 150 mm column slightly differs when compared with columns of larger diameters: In the region just above the plate the primary circulation loop forms which is reaching the inside wall of the column with the height approx. equal to 1·5 times the column diameter but this loop does reach the liquid surface. Instead, other S-shape loops proceed farther on.

Regardless of that if one loop (⌀ 300, 1 000) or other circulating loops proceeding from it (⌀ 150) form, in all cases the single-loop circulation becomes very profound. The character of this macro-circulation in bubble type columns with plates as distri-

butors can be assessed at best on basis of Table I. Without considering the qualitative nature of such results, several interesting conclusions can be drawn on their basis: *1.* Ratio of the cross-sectional area of the circulating loop is proportional to the cross sectional area of the column. *2.* The mean velocity of the liquid circulating stream is practically constant (except of the smallest gas velocities) in the whole range of measured velocities for the given compared superficial velocity. This is in agreement with the earlier published results on the effect of the size of reactors on the velocity of the upward liquid stream in the bubbled bed[3]. These velocities are, according to our measurements, larger than the published data. The difference can be explained by considerable inhomogeneity of porosity in the radial direction for our experiments which can contribute to the increase in the circulating velocity (in the original paper[3] the conditions were simulated so that all holes of the plate bubbled simultaneously and, consequently, the porosity in the radial direction was more homogeneous; moreover, according to the published radial velocity profiles the conditions in the reactor corresponded to the so-called two-loop circulation). Both these conclusions were made intuitively in our recent study and were considered to be the assumptions for derivation of relations on the effect of scaling up on mass transfer in bubbled beds[4].

It is obvious from Fig. 2 that in the considered range the diameter of the column has no effect on mass transfer. This result is of great practical importance since, generally, for design of heterogeneous bed reactors there are usually some doubts concerning the possible scaling up of reactors of laboratory sizes to industrial units. Simultaneously, it is necessary to limit our conclusion to lower beds. Only such heights were considered which were expected in individual stages of bubbled multi-

TABLE I

Cross-sectional Area of the Ascending Part of the Circulation Flow P (%) and the Liquid Velocity in This Stream u_L in Dependence on the Superficial Gas Velocity v_G and the Column Diameter D_K

V_G, ms^{-1}	D_K, m		
	0·15	0·30	1·00
0·01	$P = 8$ $u_L = 0{\cdot}4$ ms^{-1}	$P = 8$ $u_L = 0{\cdot}4$	$P = 10$ $u_L = 0{\cdot}8$
0·015	$P = 17$ $u_L = 0{\cdot}7$	$P = 14$ $u_L = 0{\cdot}6$	$P = 15$ $u_L = 0{\cdot}8$
0·02	$P = 29$ $u_L = 0{\cdot}7$	$P = 23$ $u_L = 0{\cdot}7$	$P = 22$ $u_L = 0{\cdot}8$

stage columns, with our experimental unit modelling one stage of these reactors. The insignificant effect of nonideal mixing on mass transfer rate was verified by the statistical variance of analytical data of the gas absorbed. We would like to stress that the results were obtained in the system with weak aqueous solution of electrolytes. Therefore, the possible effect of the size on changes in the structure and thus on $k_L a$ as well in concentrated electrolytes or organic liquids should not be neglected. We would like to study this problem in the near future. But it seems likely that in the measured region of column sizes from 150 to 1 000 mm the effect of column size at gas flow rates usual in this type of reactors, are the determining factor and if we assume the same degree of mixing in the compared cases it is possible to extrapolate for the fixed residence times and temperatures the overall mass transfer rate from the results obtained in the 150 mm column,

Comparison of various gas distributions was demonstrated on the 1 000 mm column. In Fig. 2 is demonstrated that both the plate and the spider type distributor give the same values of $k_L a$ though it seems by visual observation that the spider type distributor originates a better and more uniform distribution (unlike to the plate distributor, the majority of holes are active) and the mean bed porosity (in the range of accuracy of the used method) is in this arragement slightly larger. The same results obtained both with the plates (accidental bubbling of individual holes) and with the spider type distributor (forced bubbling from a larger number of holes as compared to the number of holes in the plate) has led to the conclusion that if we succeed in distributing the gas stream into the liquid in the form of bubbles without significant bypass by clusters of bubbles or without ineffective dead spaces the effect of the distributor design on $k_L a$ is obviously not substantial. For comparison, the gas was also introduced into the bed by one circular hole situated at the corner of the vessel bottom which we considered the case of extremely imperfect distribution. In this case the gas was forming large clusters of bubbles passing along the walls of the reactor, but the upper surface of the bed was bubbled in the whole cross-sectional area only in a layer about 100 mm high, while the largest portion of the reactor was not bubbled and was relatively immobile (dead space). The obtained mass transfer coefficients were in this case about half of those obtained with the distributors described above. But if we realize how extremely imperfect was the distribution of the gas in this case (when the largest portion of the reactor was ineffective), the obtained values of $k_L a$ were at these conditions relatively large. With respect to the porosity distribution along the bed height with the gas content concentrated to the upper liquid surface, we must consider just this region as the most effective from the point of mass transfer. This also demonstrates that the space in the reactor outside the circulation loops and outside the upper liquid surface is ineffective. For cases when we consider the high values of $k_L a$ (*i.e.* when the mass transfer is controlling) it is advantageous to use sectionalized bubble-type reactors, when the relative portion of the intensively bubbled bed in the region at the liquid

surface is higher in comparison to the whole volume of the reactor, than in the single stage columns.

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