

SCALING-UP OF BUBBLE-TYPE REACTORS AND THE EFFECT OF REACTOR SIZE ON GAS HOLDUP IN WATER-AIR SYSTEMS*

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On basis of assumptions on the effect of an irregular operation of sieve plates as distributors on the flow pattern of liquid in the heterogeneous bubbled bed a simplified model has been derived. The model concerns the liquid circulation on elliptical loops the type of which is dependent on the diameter of the reactor. For the model system water-air on basis of the derived model a relation has been proposed for correlation of mean porosity of the bubbled bed. The relation has been experimentally verified in columns 50, 90, 150, 300 and 1000 mm in diameter.

Scaling up of bubble-type column reactors has been studied until now only qualitatively. Here an attempt has been made to study a basical hydrodynamic parameter which is represented by a simply measurable quantity — gas holdup — in dependence on the size of the bubble-type plate reactor. On basis of our model concerning the relation between the behaviour of the bubbled bed and the operation of a bubble-type plate and on the basis of knowledge on the liquid circulation in the reactor these relations have been expressed quantitatively. The made conclusions should be useful for the study of other parameters such as interfacial area and mass transfer coefficients in dependence on the size of the reactor and for derivation of mathematical models of these types of chemical reactors.

THEORETICAL

When the gas is introduced through a perforated plate into a high bed of clear liquid (higher than 300 mm) it is possible to observe that the liquid circulates. The character of this circulation for the given reactor size and the given system is dependent on flow rates of both phases. A mathematical model¹⁻³ can be derived which describes these circulation characteristics. When actual values of parameters of this model are known (*e.g.* cross-sectional areas of circulation streams, velocity of circulation streams, velocity of bubble clusters which are identical with the circulation streams) some hydrodynamic characteristics of the bed such as the gas holdup can be calculated.

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The circulation pattern of the heterogeneous bed depends mostly on the size of the unit and on the gas flow rate which was also verified experimentally^{2,4}. By observing the circulation for different diameters of reactors and by its mathematical description the effect of the reactor size on the bed behaviour can be described. But it is obvious that 1) this approach (*i.e.* the model of pseudostationary state with mean values of characteristic quantities) requires rather time-consuming experimental measurements of the velocity and density inhomogeneity in various locations of the heterogeneous bed; 2) circulation of the heterogeneous bed is most probably the result of existence of density gradients in the bed which can occur *e.g.* by an irregular operation of the distributing plate. The point 2) suggests that the problem of effect of the reactor size on the character of the heterogeneous bed is more likely a dynamic problem. Some results on dynamic behaviour of beds with relatively low holdups (the effect of the reactor size has not been observed) support this idea⁵. In our experimental arrangement the dynamic characteristics have not been recorded and we have limited ourselves only to registration of mean values of the bed porosity.

For derivation of the dependence of porosity on the reactor size the following assumptions have been made: 1) gas entering the bed of clear liquid from the plate is bubbling, while in accordance with its accidental and changing bubbling through different holes of the plate, local irregularities in density appear in the bed; these density gradients (due to their existence there appear the local circulation loops) are the cause of pressure drop fluctuations across the bed with values of Δp_r different in different locations on the plate at the given moment. The bed is set into motion which is manifested on its surface by waves that are characterized by a single frequency or a whole set of frequencies for a given bed height, reactor size and gas flow rate. 2) In agreement with the wave motion of the liquid surface it can be assumed that the gas will most probably enter the liquid bed through those plate holes above which the mean density is at that time lower *i.e.* in places above which the minimum of wave deviations can be expected. The frequency of variation in density gradients is the greater the more uniform is the operation of the plate. 3) We assume that the wave-motion of the free surface limited by the walls of the column can be in the first approximation represented by a model of a uniformly stretched circular membrane deviated from the equilibrium state under the simplifying assumptions that the oscillations are not damped and deviations are small. The assumption 3) is not considered as too forcefull for high liquid beds. The motion of the liquid surface as of a standing wave motion in a line of points limited on both sides (if the surface is for this assumption in a section as a two-dimensional problem) is characterized by nodes at the walls of the column so that the whole column diameter must correspond to a complete number of half-waves. From the point of view of the assumption 2) there is at any moment a chance in all locations along the diameter with the exception of nodes that the corresponding space is bubbled; it is actually experimentally proved that at the walls the density of the mixture is usually the greatest and liquid flows in this region

downward from the liquid surface toward the plate (*i.e.* the two-loop circulation). The motion equation of the oscillating free surface can then be expressed by use of the wave function

$$\frac{\partial^2 z}{\partial \varrho^2} + \frac{1}{\varrho} \frac{\partial z}{\partial \varrho} + \frac{1}{\varrho^2} \frac{\partial^2 z}{\partial \varphi^2} = C^2 \frac{\partial^2 z}{\partial t^2}. \quad (1)$$

If we limit ourselves to the sine functions of time, particular solution is obtained in the form

$$z(\varrho, \varphi, t) = \exp(j\omega t) \cos(n\varphi) J_n(C\omega\varrho). \quad (2)$$

General solutions are obtained as a sum of particular solutions for all n .

The circular frequency of the oscillation is given by

$$\omega = \alpha_p C/R, \quad (3)$$

where α_p corresponds to the p -th root of the Bessel's function $J_n(\alpha) = 0$.

By analysing relations (2) and (3) for various diameters the following conclusions can be made: Position of the nodal circular curves for various diameters of columns are geometrically similar (for twice as large diameter it is situated at twice as long distances from the centre; height of waves on geometrically equivalent points (*e.g.* in the middle between two nodal curves) are for various diameters the same; the circular frequency of oscillation is indirectly proportional to the column diameter. These conclusions, especially the last one, are also valuable for qualitative considerations. The oscillation frequency is larger for smaller diameters, operation of a smaller plate is more uniform in a finite time interval, liquid above the plate is more uniformly bubbled by the passing gas. Fast "motion" of bubbling holes (in agreement with the point 2)) on the surface of a small plate causes that the initiated circulations are not expressive and that they are local and of small characteristic size. Unlike this in large columns there is a greater irregularity in their plate operation, small frequency of variation of the position of bubbling holes has a little effect on formation of macro-eddies with large characteristic dimension of the circulation loop. This quantitative model has been completely verified by the experimental observations when an expressive liquid circulation has been observed (macro-eddies) up to diameters of 300 mm and moreover, at higher linear gas velocities⁴.

Relation between the circular frequency of the surface oscillations and gas holdup can be used for description of this effect for columns of various sizes: In a finite time interval with increasing oscillation frequency increases the mean number of bubbling holes (for the given superficial velocity) related to the unit plate area (*i.e.* the actual fraction of free plate area). With increasing frequency this value of S is reaching the limiting value of S_0 (which for plates with a small number of holes and a small free plate area can be equal to the absolute number of holes N/A).

Let us assume that the simplest relation for S which satisfies the boundary conditions

$$\omega \rightarrow 0, \quad S \rightarrow 0; \quad \omega \rightarrow \infty, \quad S \rightarrow S_0 \quad (4)$$

is

$$s/A = S = S_0[1 - 1/(\omega + 1)]. \quad (5)$$

For high beds of clear liquid and for the usually used range of superficial gas velocities 0–25 cm/s the bubbles are distributed in the bubbled bed so that they form a dispersed phase and are of any possible shape (deformed bubbles have the shape irrotational ellipsoids) with the mean equivalent diameters approximately in the range of 1.5 to 6 mm (without considering some bubbles of the size of several centimeters and clusters of bubbles accidentally passing through the bubbled bed). For the mean gas porosity of the bed for the given system (under the assumption that a significant coalescence of bubbles does not take place) the relation can be written

$$\varepsilon = \bar{f} H_s \bar{V}_B s / \bar{U}_B H A, \quad (6)$$

where quantity \bar{f} which is a function of v_G and of the plate geometry, is the frequency of bubble formation from one bubbling hole, H is the mean height of the heterogeneous bed, \bar{V}_B is the mean volume of bubbles, $\bar{U}_B = \bar{U}_B(r_B)$ (plate geometry) is the mean velocity of rise of bubbles, (s/A) is the mean number of bubbling holes on a unit plate area and H_s is the actual length of path of the average bubble in the bed. For $H_s = H$ (which corresponds to the assumed plug flow of bubbles) the simplified relation holds

$$\varepsilon = \bar{f} \bar{V}_B s / \bar{U}_B A. \quad (6a)$$

The effect of irregularities of plate operation on porosity of the heterogeneous bed is formally included by combining relations (5) and (6a) and by substituting the relation (3) in the form $\omega = k/D$. We obtain

$$\varepsilon = (\bar{f} \bar{U}_B) \bar{V}_B S_0 k / (k + D). \quad (6b)$$

However, the apparently simple relation (6b) includes five parameters the values of which are doubtful. Mostly only the behaviour of a single bubble has been studied experimentally. The behaviour of clusters of bubbles at their massive formation is different from that of a single bubble and reliable experimental data on them are mostly missing. The quantity S_0 has been determined neither directly nor experimentally. The explicit expression of porosity by the relation (6b) as a function of linear gas velocity and of the column diameter is thus practically impossible.

The dependence of the gas porosity on linear gas velocity for the given column diameter has been already derived on the basis of considerations concerning the macroflow of the gas passing through the bubbled bed and can be now used for expression or substitution of unknown quantities in the relation (6b) so that we write

$$\varepsilon = \beta v^{4/5} / (2v + 20)^{7/15}, \quad (7)$$

where β is a function of only physico-chemical properties and of the size of the unit. In agreement with the relation (6b) the general size of the unit can be written ($S_0 = \text{const.}$)

$$\beta = \gamma k / (k + D), \quad (8)$$

and the final relation for the gas holdup is

$$\varepsilon = \gamma [k / (k + D)] [v^{4/5} / (2v + 20)^{7/15}], \quad (9)$$

where quantities γ and k can be obtained for the system water-air experimentally.

In agreement with the considerations made in our previous study³ it is in general possible to consider the quantity k , for the systems of non-electrolytes and without the chemical reaction between the gas and liquid, a universal constant, while the quantity γ is a function of physico-chemical macro-quantities of the system.

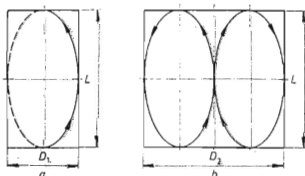


FIG. 1

Paths of Bubbles Approximated by Ellipses
a Single-loop circulation on the ascending half-ellipse, b two-loop circulation.

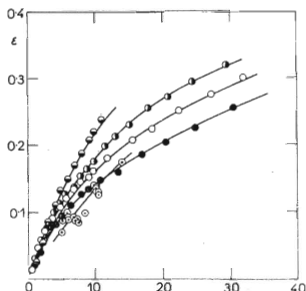


FIG. 2

Effect of Reactor Size on Porosity of the Bubbled Bed for the Air-Water System

● D 50 mm, ● D 90 mm, ○ D 150 mm,
● D 300 mm, ○ D 1000 mm. v in cm s^{-1} .

The dependence of porosity on the diameter of the column and on the linear gas velocity can be without difficulties expressed by relation (9) where we have assumed that $H_s = H$ up to such sizes of column reactors where a significant effect of curvature of the bubble path does not appear. In agreement with our considerations on irregular operation of the plate it is obvious that with reactors of large sizes the initiated macro-eddies cause that the actual length of the bubble path is $H_s > H$. The porosity ε is (for $v = \text{const}$) given as the result of superposition of two counter-acting factors which are expressed by the relation $k/(k + D)$ which results from the model on irregular operation of plates with increasing size and which acts in the direction of decreased porosity and of the factor (H_s/H) that becomes significant just for large reactor sizes and which, on the contrary, contributes to the increase in porosity of the heterogeneous bed in comparison with reactors of smaller diameters at otherwise same conditions. For derivation of the ratio H_s/H the following assumptions are made: 1) Bubbles or clusters of bubbles are moving in the bubbled bed on elliptical paths and at the increase of the linear gas velocity the flow pattern of the liquid mixing is being transformed from the one to the two-loop circulation. 2) One characteristic dimension of the circulation loop which is forming the path of bubbles is comparable with the size of the experimental unit where actually the horizontal axis for a single-loop circulation equals to $D/2$ and for the two-loop circulation to $D/4$ (Figs 1a and 1b). 3) Height of the elliptical circulation loop L is constant and thus independent on the diameter of the unit and on the height of the heterogeneous bed. We assume along the height of the reactor the existence of the elliptical (H/L) circulation loops (one or two-loop). 4) In agreement with the experimental results on the effect of the reactor diameter on the ratio (H_s/H) the limits must hold

$$\begin{aligned} \lim_{D \rightarrow 0} (H_s/H) &= 1 \text{ (for an arbitrary } v \text{);} \\ \lim_{D \rightarrow \infty} (H_s/H) &= f(D, v). \end{aligned} \quad (10)$$

For assumptions 1) to 4) there exists an experimental foundation. The successive change of circulation loops at the increase in the linear gas velocity was quite positively observed and up to the sizes that we have experimentally studied *i.e.* up to the diameter of 1 m the approximation according to point 2) can be accepted. The elliptical path, unlike that of Reith⁶, who assumes the rise of bubbles on circular paths, is a more natural expression of the effect of the column diameter on the circulation loop. For the one-loop circulation the bubbles are usually following the upward path of the half-ellipse and are for low gas velocity not entrained into the downward elliptical path directed toward the distributing plate. Then at the increased gas velocity the two-loop circulation begins to predominate so that the gas is passing in the upward stream whose location of origin is not fixed (it does not need to be only in the plate centre) and the clear liquid circulates downward at the walls. By the increase of the gas flow rate the bubbles also appear in the downward streams. The simplest geo-

metrical approximation of this complex mechanism of mixing, which is in a qualitatively good agreement with the exact situation, lies in description of the two-loop circulation by two symmetrical ellipses. Assumption 3) has been verified only for the relatively low ratios (H/D) especially for large sizes of reactors. We estimated the height of the macro-eddy (on which are usually superposed much smaller secondary eddies) independently on the size of the equipment within the range from 700 to 1000 mm. The assumption of constant eddy height (circulation loops) for the given reactor size is then in agreement with the found experimental result according to which the mean porosity does not substantially change with the height of the heterogeneous bed. Finally, with assumption 4) is practically introduced the correlation for the effect of the reactor size on deviation of the actual path of bubbles from the direct motion (*i.e.* the effect of one or two-loop circulation on H_s/H for small diameters is negligible).

The circumference of the ellipse of height L and width D can be approximated by the relation

$$H_s = 2\pi[1/2(D^2/4 + L^2/4)]^{1/2}, \quad (11)$$

where we assume for the one-loop circulation that the bubbles move on the half-ellipse, with the relative path length given by

$$H_s/H = H_s/L = (\pi/2 \sqrt{2}) (D^2/L^2 + 1)^{1/2}. \quad (12)$$

In the second limiting case the bubbles move at the two-loop circulation along the fully developed paths of two ellipses. In this case also the backmixing of the gas appears in the bed and the relative length of the path is given by

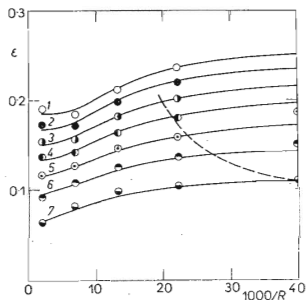


FIG. 3

Theoretical Dependence of Porosity on Radius of Reactor R (mm) and Experimental Data

Solid line Eq. (16); dashed line is the lower limiting gas velocity at which plug-flow appears; experimental points: 1 $v = 0.16 \text{ m s}^{-1}$, 2 0.14, 3 0.12, 4 0.10, 5 0.08, 6 0.06, 7 0.04.

$$H_s/H = (\pi/\sqrt{2})(D^2/L^2 + 4)^{1/2}. \quad (13)$$

According to the assumption 1) with increasing gas velocity both types of circulation develop continuously *i.e.* in general the relation holds

$$\frac{H_s}{H} = \frac{\pi}{\sqrt{2}} \left[\left(1 - \frac{3}{4+v} \right) \left(\left(\frac{D}{L} \right)^2 + 4^{1-1/(1+v)} \right) \right]^{1/2}, \quad (14)$$

where for $v \rightarrow 0$ the relation (12) and for $v \rightarrow \infty$ the relation (13) is obtained. For practical values of v and D the general relation (14) can be applied, where according to assumption 4) the extent of the effect of relation (14) on the value of porosity is dependent on the size of the unit *i.e.* the relation (9) must be multiplied by the effectivity coefficient

$$a = a[(H_s/H), D]. \quad (15)$$

EXPERIMENTAL

Procedure. In this study, we have been mainly concerned with the system water-air where the unfavourable effects like with electrolytes (foaming) or such types of gas-liquid systems which together mutually react (the effect of interfacial turbulence on the porosity of the bed) do not appear. The measurements were performed in single stage columns of diameters 50, 90, 150 and 300 mm of circular cross-sectional area, while the column of 1000 mm had a square cross-section. The air has been supplied by the central system of pressurized gas and for the 1000 mm column from the system of gas blowers. Distilled water and tap water (column 1000 mm) have been used. Air was introduced into the distributing chamber below the distributing plate, then passed through the grid with relatively small free area which was situated below the distributing plate and was in contact with the liquid.

Plate parameters and distributing chambers: column 50 mm: Distributing plate $d = 5$ mm, $\varphi = 5.94\%$, size of chamber below the internal grid $V_1 = 785 \text{ cm}^3$, volume of the chamber between the grid and distributing plate $V_2 = 196.25 \text{ cm}^3$, free plate area of the internal grid $\varphi_2 = 1\%$. Column 90 mm: Distributing plate $d = 5$ mm, $\varphi = 5.94\%$, $V_1 = 2543.4 \text{ cm}^3$, $V_2 = 635.85 \text{ cm}^3$, $\varphi_2 = 1\%$. Column 150 mm: $d = 5$ mm, $\varphi = 5.94\%$, $V_1 = 7065 \text{ cm}^3$, $V_2 = 1766.3 \text{ cm}^3$ and 7065 cm^3 , $\varphi_2 = 1\%$. Column 300 mm: $d = 5$ mm, $\varphi = 3\%$, $V_1 = 28260 \text{ cm}^3$, $V_2 = 7065 \text{ cm}^2$, $\varphi_2 = 1\%$. Column 1000 mm: $d = 3$ mm, $\varphi = 3\%$, $V_1 = 256000 \text{ cm}^3$, $V_2 = 344000 \text{ cm}^3$, $\varphi_2 = 1\%$.

Porosity: From preceding studies has resulted that the plate geometry has no effect on the value of mean porosity. The bed heights were chosen 1200 mm and in the column 1000 mm the heights 600 and 900 were studied. The bed height has no effect on mean porosity (for $h_{\min} > 200$ mm). The size of the chamber between the plate and the grid with small free plate area has no effect on the value of porosity. The porosity of gas has been measured by the method described earlier⁶. The mean (integral) value of quantity $\bar{\epsilon}$ has been correlated. The dependence of the mean gas porosity in the bubbled bed on linear gas velocity and on the column diameter is given in Fig. 2.

RESULTS

For the system water-air were determined the following values of constants $\gamma = 0.1925$, $k = 45.6$. The resulting general relation for porosity of the bubbled bed for this system is

$$\varepsilon = 0.1925 \left(\frac{45.6}{45.6 + D} \right) \left(\frac{v^{4/5}}{(2v + 20)^{7/15}} \right) \left\{ 1 + \frac{\pi/\sqrt{2} \{ [1 - 3/(4 + v)] (D/100)^2 + 4^{1-1/(1+v)} \}}{1 + 1/[0.291 (D/100)^2]} \right\}. \quad (16)$$

The dimensions of quantities are as follows: $[D] = \text{cm}$, $[v] = \text{cm s}^{-1}$, $[\varepsilon] = [-]$. In Fig. 3 theoretical curves of porosities for various superficial gas velocities are plotted as the parameter, in dependence on the reactor diameter and the experimental data from Fig. 2. The agreement of the experimental and theoretical results is very good. By the proposed procedure has been, to a certain extent, theoretically expressed also the unexpected dependence of porosities on the diameter and on linear gas velocities for large diameters. But at the same time also becomes obvious the necessity of experiments with reactors having the diameter $D > 1000 \text{ mm}$ so that validity of the dependence (16) could be extended. The smallest column diameter which can be modelled is 90 mm. In agreement with one of our assumptions made for the derivation, we have assumed that the gas is present as the dispersed phase, the proposed correlation is valid up to such combination of reactor diameters and gas velocities when gas bubbles of the size covering the whole diameter of the reactor do not form. From our correlation it is obvious that the diameter 50 mm significantly differs from the others because here the so called effect of walls becomes significant. Though the proposed procedure seems to be hopeful it is not possible at present to estimate the behaviour of the heterogeneous bed in larger diameters of reactors despite our intention to obtain a general correlation. The diameter 1000 mm differs so much from the next smaller diameter 300 mm that final conclusions cannot be made. The experiments with reactors of large diameters are thus desirable though they are far outside the scope of ordinary laboratory practice. But it is not possible to accept the frequently made statement that modelling in columns of the diameters about 300 mm is sufficient for scaling-up of bubble type reactors. Our results verified up to the height of the heterogeneous bed 1200 mm are, with respect to the practical arrangement of bubble-type reactors as multistage columns (where the height of the stage is usually equal to the diameter of the reactor), sufficient for verification of the behaviour of the heterogeneous bed.

LIST OF SYMBOLS

a	effective coefficient for the effect of reactor size, Eq. (15)
A	area of distributing plate
C	velocity of wave motion in liquid
D	reactor diameter
d	diameter of holes of the distributing sieve plate
T	mean frequency of bubble formation from one bubbling hole
H	mean height of the heterogeneous bed above the distributing plate
H_s	actual length of bubble path in the bed
j	unit vector
$J_n(\alpha)$	Bessel function
k	constant in relation (6b)
L	vertical dimension of the circulation loop
Δp_f	pressure drop across the bubbled bed
R	radius of reactor
s	number of bubbling holes
S	number of bubbling holes per unit of plate area
t	time
v	superficial (linear) gas velocity
\bar{V}_B	mean bubble volume
\bar{U}_B	mean velocity of bubble rise
z	deviation
α_p	p -th root of the Bessel function
β	function
γ	constant
ϵ	gas porosity
φ	coordinate
φ	free area of distributing plate
φ_2	free area of internal grid
ω	circular wave frequency
q	coordinate

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