

GAS HOLDUP AND ITS RELATION TO INTERFACIAL AREA IN BUBBLE-TYPE REACTORS

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A relation has been derived and experimentally verified for calculation of the gas holdup in bubble-type plate reactors of the gas-liquid type. The effect of physico-chemical properties of the system and of the geometrical parameters on gas holdup are discussed. The proposed relation is suitable for prediction of the interfacial area on basis of the known gas holdup, since experimental determination of the interfacial area is complicated and very frequently disputable.

Reactors of the bubble-type are suitable for slow reactions between the gas and liquid when the rate of absorption with the chemical reaction is only controlled by the reaction rate in the bulk of liquid and thus when it is directly proportional to the liquid present in the reactor. Liquid holdup in the reactor is in this case practically the only one, though a very important, hydrodynamic parameter determining the size of the reactor. This study is concerned with the gas (and/or liquid) holdups in bubble-type reactors when the gas is distributed into the liquid by a sieve plate with circular holes and when the reactor, as concerns the liquid flow, is a batch one. In practical applications the liquid flow rate is usually very low and its effect on gas holdup is negligible¹. Thus our studied system is a suitable approximation also for reactors with both phases in continuous flow. Liquid holdup and interfacial area in bubble-type reactors has been studied lately by Reith². This study is of considerable importance as it presents a simple universal relation for the gas holdup on basis of considerations concerning the motion of gas bubbles; but in a reactor it does not represent the actual conditions and it does not include the effect of physico-chemical properties of various systems which is indisputable.

It can be expected that in a bubbled bed there will exist a direct dependence between the gas holdup and the interfacial area. Interfacial areas will depend on the gas flow rate, height of the bubbled liquid bed and its width *i.e.* on parameters affecting the size of bubbles and the character of their motion in the reactor. It will also be in general considerably dependent on physico-chemical properties of the system but especially on the effects in vicinity of the interfacial area which are affecting the structure of the bubbled bed (*e.g.* presence of particles with an electrostatic charge affecting among others the coalescence of bubbles, or traces of surface active matters affecting foamibility *etc.*). These effects may be considerably different for various systems without the basical macroparameters by which the physico-chemical properties of the system are usually characterised such as the density, viscosity and interfacial

area, being expressively altered. Profound changes in structure of the heterogeneous bed may take place as the gas reacts with liquid. The effect of structure of the heterogeneous bed and the distribution of bubble sizes then affects not only the gas holdup in the unit of the given size but scalling-up as well when in different systems even a completely reversed dependence of gas holdup on dimensions of the unit can be found. The effect of the discussed phenomena at the interface on structure of the heterogeneous bed cannot be quantitatively expressed and as regards determination of the holdup in a given system we shall have to rely in design of gas-liquid reactors on direct experimental measurement in a laboratory size especially when electrolytes are concerned. But it is possible to expect that there exist systems where relations for liquid holdup can be derived with the use of physico-chemical macroquantities. Such systems are nonelectrolytes in contact with gas without chemical reaction between the gas and liquid. They are very frequently used as the "classical" testing systems (*e.g.* water-air). However, the results obtained with such systems enable only an estimate of behaviour of actual systems. Similarly for determination of the interfacial area by a chemical method the found values are valid only for the measured system (usually electrolyte) in which the found interfacial area corresponds to a certain gas holdup. The results cannot be easily applied to another system, in spite of the same or similar values of the physico-chemical macroquantities. But it is possible, without regard to the character of the used liquid, to derive a general dependence between the gas holdup and the interfacial area. By measuring one of these hydrodynamic parameters it is possible from such a dependence to estimate the second one which is of special importance in the case of interfacial area the experimental determination of which is incomparably more difficult than that of gas holdup. Here, we are deriving a relation between the gas holdup and the interfacial area and we are analysing it for the case when the liquid is an electrolyte or nonelectrolyte.

THEORETICAL

The pressure drop of a gas bubbling through a bed of liquid can be formally expressed by the relation for pressure drop in a pipe having an equivalent diameter d_e by

$$\Delta p_G/l = \frac{1}{2}(\psi_G/\rho_G)(1/d_e)(G/e)^2, \quad (1)$$

where the equivalent diameter of the pipe is expressed from the ratio of gas holdup and interfacial area, with both quantities related to the volume of the bubbled bed in the reactor according to

$$[(\pi d_e^2/4) l/V_R]/(\pi d_e l/V_R) \equiv e/a = d_e/4. \quad (2)$$

For a heterogeneous bed without liquid flow this pressure drop of gas across the bed

equals to the weight of the liquid bed

$$\Delta p_G/l = \rho_L g . \quad (3)$$

A simple reaction has been proposed by Reith² according to which the interfacial area is proportional to the gas velocity according to relation

$$a = K[v/(2v + 0.2)] , \quad (4)$$

where K is dependent in general on properties of the liquid and gas and on dimensions of the reactor. For $G = \rho_G v$ by combining relations (1) to (4) we obtain

$$e = [(\frac{1}{2})(1/g)(\rho_G/\rho_L)K]^{1/3} \psi_G^{1/3} [1/(v + 0.2)^{1/3}] v . \quad (5)$$

The equivalent Reynolds number Re_e for the gas flow rate through the bubbled bed is defined

$$Re_e \equiv v d_e \rho_G / \mu_G . \quad (6)$$

In the region of usual linear gas velocities in bubble-type reactors $v = 0.01$ to $0.2 \text{ m} \cdot \text{s}^{-1}$ the equivalent diameter is in the range from 0.0025 to 0.004 m (water-air) and with the corresponding range of Re numbers $Re_e = 1.6$ to 54 . In agreement with the formal analogy of bubble flow through the bubbled bed with gas flow through a pipe in laminar flow it is possible to expect that the friction factor can be expressed by a simple function of the Reynolds number. We assume the validity

$$\psi_G = D/Re_e^n , \quad (7)$$

where n in general need not be equal to one, but in agreement with the analogy it should not differ much from this value. By use of relations (6) and (7) for gas holdup we may write

$$e = (1 - e_L) = \frac{[(1/g)(\rho_G/\rho_L)]^{1/(n+3)} D^{1/(n+3)} (\mu_G/\rho_G)^{n/(n+3)} K^{(1+n)/(n+3)} v^{3/(n+3)}}{2^{3/(n+3)} 4^{n/(n+3)} (2v + 0.2)^{(n+1)/(n+3)}} . \quad (8)$$

Values of quantities D and n may be found experimentally and they should be for the used range of gas flow rates universal. Values of the constant K can be obtained from the experimentally determined interfacial area.

EXPERIMENTAL

Gas holdup for selected gas-liquid systems was measured with columns having diameters 150 and 300 mm. The controlled gas entered the distributor at the bottom of the column and was distributed into the static liquid bed by sieve plates. Plates with circular holes situated in edges of equilateral triangles were used. Hole diameter varied in the range of 1.6 to 5 mm, free plate area 0.5 to 18%. The initial heights of static liquid bed were in the range of 600 to 1200 mm, linear gas velocities 0 to 30 cm/s. Gas (liquid) holdup was measured by the method of pressure differences based on measurements of the static pressure along the bed height^{1,2-4}. The mean gas holdup, which was correlated, was calculated as the integral of the ratio of pressure differences and distances of pressure taps over the whole bed height. The number of pressure taps along the bed height was sufficient for a safe description of holdup distribution inside the bubbled bed.

RESULTS

The System Electrolyte-Gas

Relation (8) can be applied to the system of aqueous solution of sodium sulphite-air columns of 150 and 300 mm diameter. The effect of geometry of the sieve plate in the range $d = 1.6$ to 5 mm, $\varphi = 0.53$ to 17.96% and liquid heights 600 to 1200 mm has been found earlier⁴ with the system water-air as insignificant (Fig. 1). The sulphite solution for verification of relation (8) was chosen because there exist safe methods for measurements of the interfacial area by the sulphite method and the coefficient K from these experiments can be used directly. At first the value of exponent n was looked for according to the simplified relation (8) in the form $e_{\text{exp}} = \text{const. } f(v)$. The value $n = 3/4$ was found to be best fitting the experimental points (Fig. 2). The relative deviation of calculated holdups from those experimentally determined is $\pm 3.25\%$, with the greatest deviation encountered in the region of low linear gas velocities. The agreement of the experimentally determined holdup values with the calculated ones is very good and relation (8) completely expresses the shape of the dependence of gas holdup on gas velocity. The obtained dependence was then used for calculation of the dependence of gas holdup on gas velocity for the same system in the column with 300 mm diameter. Under assumption that the value of the constant D is universal, the ratio of holdups in both columns is for $v = \text{const}$ given by ratio of K values. In our case $e_{300}/e_{150} = (1675/961)^{7/15}$ where values of constants K were obtained as slopes of straight lines passing through the points from Fig. 6.11 of the study made by Reith². The exponent 7/15 was obtained from the theoretical relation (8) for the above determined exponent 3/4. For e_{150} values taken from the curve 1 were used. Then, $e_{300} = 1.296e_{150}$. It is obvious from Fig. 2 that the calculated curve is satisfactorily fitting the shape of the experimentally determined dependence in spite of difficulties with determination of the accurate values of K from the graph. This conclusion is important as it confirms our assumption concerning the validity of dependences relating the interfacial area and gas holdup in the

presented form and it also demonstrates that the same relation (8) will be valid for different sizes of columns. It is necessary to realize that scaling-up cannot be studied without taking into consideration the structure of the heterogeneous bed as it is expressed through the constant K . We are not aware of any other method, than the direct experimental measurement, according to which the constant K could be calculated for any gas-liquid system and neither how it varies with increasing size of the unit. It is known that in bubble columns circulation loops form in which bubbles follow the motion of liquid. The character of circulation and the structure of the heterogeneous bed are mutually related and can differ from system to system. As the character of circulation is affected by the size of the unit, various systems can behave differently at increasing their size. In Fig. 1 are plotted experimental gas holdups for the system water-air for comparison of columns of 150 and 300 mm diameter. It is obvious that the dependence of gas holdup on the dimension of the reactor is completely reversed to that in the system aqueous sulphite solution-air while the

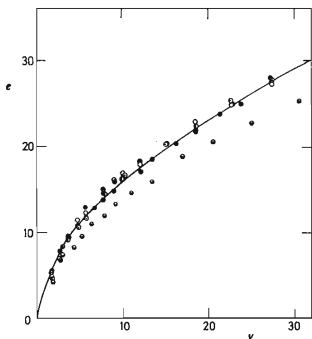


FIG. 1

Dependence of Gas Holdup (e) on Linear Gas Velocity for the Water-Air System

Column diameter 150 mm: \circ d 1.6 mm, φ 6.02%, h 1200 mm; \bullet d 1.6 mm, φ 0.53%, h 1200 mm; \otimes d 5 mm, φ 17.96%, h 1200 mm; \ominus d 5 mm, φ 5.94%, h 1200 mm; \odot d 1.6 mm, φ 6.02%, h 600 mm. Column diameter 300 mm: \ominus d 5 mm, φ 3.04%, h 1200 m.

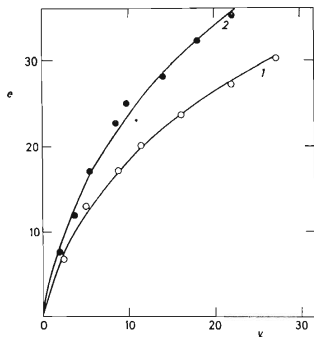


FIG. 2

Comparison of Calculated and Experimental Holdups for the 0.8 mol/l Aqueous Solution of Sodium Sulphite Air

1 Calculated gas holdup from Eq. (8) for $n = 3/4$, column 150 mm. 2 Calculated gas holdup from curve 1 and ratio of K values for the column 300 mm. Experimental data: \circ d 1.6 mm, φ 6.02%, h 600 mm, column diameter 15 mm; \bullet d 1.6 mm, φ 3.04%, h 600 mm, column diameter 300 mm.

macroproperties of both systems do not differ considerably. For behaviour of individual systems with scaling-up no quantitative method is available. At present neither the structure of the bed characteristic for the given diameter of the unit can be relevantly determined. This is clearly demonstrated in Fig. 3 where sulphite solutions were bubbled by air or oxygen. The dependence of gas holdup on linear gas velocity for systems sulphite-air and sulphite-oxygen are nearly identical. For the system sulphate-oxygen, in agreement with the relation (8), the expected dependence $e-v$ can be derived *e.g.* from the similar dependence of the system sulphite-air under the assumption of equal K values. In Fig. 3 are plotted two different sets of experimental points for the system sulphate-oxygen. The theoretical curve is then situated in the experimentally obtained range of holdup close to the lower values, which are close to the behaviour of the system sulphate-air at otherwise same conditions. The differences obtained for the system sulphate-air can be explained *e.g.* by the presence of surface active matters in air taken from the pressure vessels which may vary. But the most probable explanation can be based on Fig. 4 from which is obvious that the process of formation of a very fine-dispersion of oxygen in sulphate solutions is dependent on time and is slowly steadying while the experimental gas

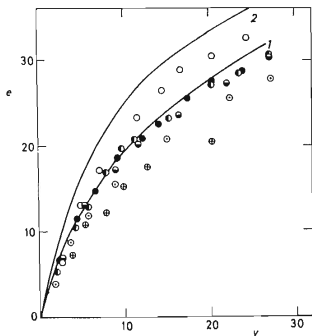


FIG. 3

Gas Holdup for the System Electrolyte-Gas in the Column 150 mm in Diameter

○, ● 0.8 mol/l Na_2SO_4 -oxygen; ● 0.8 mol/l Na_2SO_4 -air; ⊗ 0.8 mol/l Na_2SO_3 -oxygen; ⊙ 0.8 mol/l Na_2SO_3 -air; ⊖ water-air.
1 Theoretical $e-v$ dependence for the 0.8 mol/l Na_2SO_4 -oxygen; 2 dependence $e-v$ for electrolytes according to Reith².

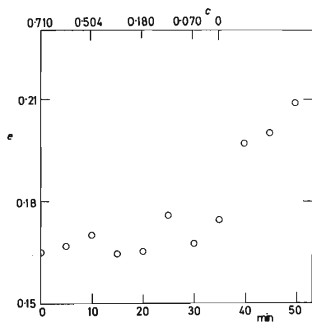


FIG. 4

Variation of Oxygen Holdup with Time in Aqueous Soln. of Sodium Sulphite at its Oxidation to Sulphate

v 0.96 ms^{-1} , h 600 mm, $(c_{\text{SO}_3^{2-}})_{\text{initial}} = 0.71$ mol/l.

holdups for the given gas velocity are in the range of experimental data presented in Fig. 3. This system is more likely to form stable foams, the formation of which is at the beginning dependent on time. This tendency was observed as well in the system sulphite-air, though on a much limited scale. Thus the experimental procedure is affecting as well values of the experimentally determined gas holdups. It is obvious that for these cases equal values of the constant K for both considered systems cannot be assumed (Fig. 4). In measurements of gas holdups in the system sulphite-air at a constant gas velocity took also place oxidation of sulphite to sulphate, nevertheless the gas holdup and structure of the bed remained practically constant till the complete conversion of sulphite even at a large excess of sulphate ions. When the oxidation is completed, the gas holdup considerably increases. There are two

TABLE I
Properties of Electrolyte Solutions

Aqueous soln. of	Gas	Concentration mol/l	σ dyn/cm	μ_L cP	ϱ_L g/cm ³
Na ₂ SO ₃	air	0.73	70.09	1.23	1.0254
		0.61	66.40	—	—
		0.46	64.82	—	—
		0.32	61.66	—	—
Na ₂ SO ₄	air	0.8	64.60	1.21	1.0283
		Na ₂ SO ₃	O ₂	0.74	52.70
Na ₂ SO ₄	O ₂	0.66	52.17	—	—
		0.52	52.7	—	—
		0.8	62.19	—	—

possibilities how to explain that state: 1) the presence of traces of sulphite is affecting the physico-chemical properties of the liquid with the corresponding change of the bed structure. The changed macroquantities (ϱ_L , μ_L , σ) need not be significant and the sulphite ion are acting as the surface active matter *i.e.* differently than the sulphate ion. Physical properties of both systems are given in Table I. But because both these systems behave practically in the same manner with air, this possibility does not seem to be justified. 2) At bubbling the sulphite solution by oxygen, the oxidation is much more active than in the case of bubbling by air. Mass transfer across the interface is considerably affecting the phenomena on the phase boundary with its size different for different systems. Regardless of probability of these explanations it is obvious that in systems with electrolytes once upon the time different behaviour

may be expected which at present cannot be expressed by a correlation relation *i.e.* by giving the value of K . An experimental study of a number of systems of electrolytes as well as reactions of gas with liquid is needed on the basis of which at least some groups of systems with similar behaviour can be defined which is true from the view of scaling-up as well.

The System Nonelectrolyte-Gas

For verification of relations (8) for the system nonelectrolyte-gas measurements of gas holdups in systems water-air, aqueous solution of glycerol-air and ethyl alcohol were made. The values of constants K were calculated according to relation (4) where for the interfacial area was substituted the value calculated from the relation given by Calderbank³ for sieve plates

$$a = 0.38 \left(\frac{v}{26.5} \right)^{0.775} \left(\frac{v p_L}{(N_0/A) d \mu_L} \right)^{0.125} \left(\frac{\rho_L g}{d \sigma} \right)^{1/3}, \quad (9)$$

where $a = \text{cm}^{-1}$, $v = \text{cm s}^{-1}$, $\rho_L = \text{cm}^{-3} \text{g}$, $\mu_L = \text{g cm}^{-1} \text{s}^{-1}$, $\sigma = \text{dyn cm}^{-1}$, $d = \text{hole diameter, cm}$ and N_p/A is the number of holes per unit of plate area. Calderbank has verified this relation for water, alifatic alcohols and glycol with the largest used diameter of the column 132 mm. As Calderbank is not taking into account the change of interfacial area at scaling-up, the relation (9) has been applied only to our column of 150 mm in diameter.

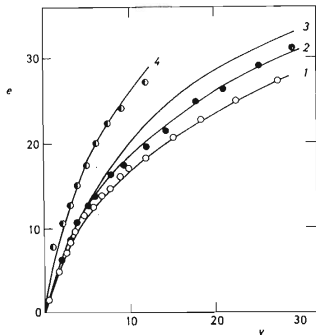


FIG. 5

Air Holdup in Selected Systems of Nonelectrolytes in the Column 150 mm in Diameter, h 600 mm

○ Water-air, d 1.6 mm, ϕ 6.02%; ● ethanol-air, d 3.0 mm, ϕ 2.96%; ● aqueous soln. of glycerol-air, d 3.0 mm, ϕ 2.96%. 1 Water-air, $K = 750$; 2 ethanol-air, K 840; 3 dependence according to Reith² for nonelectrolytes, 4 solution of glycerol-air, K 2212.

System Water–Air

Dependence of calculated values of a according to relation (9) on parameter $v/(2v + 0.2)$ is not, like in the study of Reith, straight but nevertheless it can be approximated by a straight line up to linear gas velocities 10 cm s^{-1} and the mean value of K can be determined. The found value of K for the system water–air in the column of 150 mm diameter is $K = 750$. The shape of the expected dependence $e-v$ can be derived by use of relation (8) in the simplest way from the known ratio of constant K of two systems. In this case *e.g.* the dependence sulphite–air can be used for which at the given linear velocity compared the relation $e_{\text{SO}_3}/e_{\text{H}_2\text{O}} = (961/750)^{7/15}$, holds under assumption of practically equal densities of both liquids). In Fig. 5 is obvious an excellent agreement of the experimental points with the calculated curve. The mean deviation is $\pm 1.9\%$ in the whole range up to 30 cm s^{-1} .

The System Ethylalcohol–Air

Theoretical curve of dependence $e-v$ has been derived from the relation for the system water–air according to the relation $e_{\text{C}_2\text{H}_5\text{OH}}/e_{\text{H}_2\text{O}} = (\rho_{\text{L,H}_2\text{O}}/\rho_{\text{L,C}_2\text{H}_5\text{OH}})^{4/15} (K_{\text{C}_2\text{H}_5\text{OH}}/K_{\text{H}_2\text{O}})^{7/15}$. The value of K for the system ethyl alcohol–water has been determined from the dependence of interfacial area on velocity according to relations (4) and (9), as in the last example. In this case $K = 840$. The agreement of experiments with the calculated dependence is given in Fig. 5.

The System Aqueous Solution of Glycerol–Air

The mean value of constant K has been calculated for this system from the experimental dependence $e-v$: $e_{\text{g}}/e_{\text{H}_2\text{O}} = (\rho_{\text{L,H}_2\text{O}}/\rho_{\text{L,g}})^{4/15} (K_{\text{g}}/750)^{7/15}$. The value of the mean constant is 2212. For bubbling of air through glycerol a finely dispersed foam forms. The agreement of the theoretical curve with the experimental dependence is obvious from Fig. 5.

The General System Nonelectrolyte–Gas

On basis of the above given experimental results an empirical relation was calculated for the constant K of nonelectrolytes

$$K = C\mu_{\text{L}}^{0.23}/\sigma^{0.077}, \quad (10)$$

where $[\mu_{\text{L}}] = \text{cP}$, $[\sigma] = \text{dyn/cm}$ with the value of the constant $C = C_1 f$ (diameter of the column) = 1042 for the column diameter 150 mm. By combining relations (4) and (10), the interfacial area for the system nonelectrolyte–gas can be estimated for the given diameter of the reactor. The mean deviation of calculated values of inter-

facial areas according to relations (4) and (1) from the values calculated according to Calderbank (relation (9)) in the region of linear gas velocities $v = 0-15 \text{ cm s}^{-1}$ is less than 18%. The relation (10) was verified for the range of parameters of the liquid $\rho_L (0.789-1.196 \text{ cm}^3)$, $\mu_L (1-107.4 \text{ cP})$, $\sigma (22.3-73.0 \text{ dyn cm}^{-1})$.

Friction Factor ψ_g

Theoretical dependences of gas holdups on linear gas velocity have been so far obtained by comparison with an arbitrarily chosen reference system. The assumption concerning the independence of the constant D on the system and the reactor size has thus been, due to a good agreement of experimental and theoretical $e-v$ dependences, confirmed. So relation (8) can be directly used for calculation of theoretical gas holdups while the mean value of the constant D has been calculated from relation (11) valid for the water-air system into which experimental values and the respective physico-chemical parameters have been substituted. From this relation

$$e = \frac{1}{2^{4/5}} \frac{1}{4^{1/5}} \left(\frac{1}{g} \frac{\rho_G}{\rho_L} \right)^{4/15} D^{4/15} \left(\frac{\mu_G}{\rho_G} \right)^{1/5} K^{7/15} \frac{v^{4/5}}{(2v + 0.2)^{7/15}} \quad (11)$$

the value $\bar{D} = 1.74 \cdot 10^3$ has been obtained.

Figs 4 and 5 also illustrate the dependence $e-v$ according to Reith, *i.e.* in accordance with the relation $e = v/(2v + b)$ where for electrolytes $b = 0.2$ and for nonelectrolytes $b = 0.3$. Our experimental $e-v$ data obtained with actual systems are not described by this simple universal relation while the deviations are considerable. But the shape of curves calculated from the relation by Reith is similar to those experimental, and thus this relation can be used, due to its simplicity, as the first approximation for calculation of gas holdups.

LIST OF SYMBOLS

a	specific interfacial area (m^{-1})
d	hole diameter of the sieve plate (mm)
d_e	equivalent diameter (m)
D	constant in relation (7)
e	relative gas holdup (porosity)
$G = \rho_G v$	mass flow rate of gas ($\text{kg m}^{-2} \text{ s}^{-1}$)
g	gravitational acceleration (m s^{-2})
h	clear liquid height on plate (m)
K	constant in Eq. (4)
l	equivalent pipe length (m)
Δp_G	gas pressure drop in bubbled bed (N m^{-2})
$\text{Re}_e = v d_e \rho_G / \mu_G$	equivalent Reynolds number
v	linear gas velocity (ms^{-1})

V_R	volume of bubbled bed in reactor (m^3)
ψ_G	friction factor
ρ_G, ρ_L	gas (liquid) density ($kg\ m^{-3}$)
μ_G	gas viscosity ($kg\ m^{-1}\ s^{-1}$)
φ	free plate area
σ	interfacial tension

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