

GAS HOLD-UP IN AGITATED AQUEOUS SOLUTIONS OF STRONG INORGANIC SALTS*

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The over-all hold-up of gas dispersed in a mechanically agitated liquid is strongly influenced by the presence of dissolved inorganic salts. The properties of the dispersion cannot be adequately described by the commonly studied physical properties of liquid, *i.e.*, the density ρ , the viscosity η and the surface tension σ . Lee and Meyrick have shown that, to describe the gas hold-up in electrolytic solutions, a new quantity can with advantage be introduced

$$\psi = c(d\sigma/dc)^2 \phi,$$

where c is molar concentration and ϕ is defined by

$$\phi = (1 + d \ln f/d \ln c)^{-1},$$

where f is the activity coefficient. We measured the gas hold-up in various electrolytes at a number of concentration, impeller rpm and gas flow levels. With increasing value of the quantity the gas hold-up increased up to a certain value and then remained constant.

The gas hold-up was correlated with the quantity ψ , the gas flow, and the Weber number, yielding a dimensionless equation with constants depending on the geometry of the apparatus.

The problems connected with the search for regression of semiempirical formulas describing gas hold-up in an agitated batch were investigated in a number of original papers; however, most of the work was limited to cases where water was used as the liquid phase. Minor attention was paid to studies into the problem of engineering importance: how the hold-up is affected by the physical properties of the liquid. These studies again must be divided into two groups, the one using organic species or their aqueous solutions for the liquid phase and the other dealing with hold-ups in electrolyte solutions, mostly in aqueous solutions of inorganic salts.

Calderbank¹ followed by other authors^{2,3} have shown that a single relationship cannot suffice for an adequate description of the behaviour of gas both in aqueous electrolyte solutions and in non-electrolytes, *e.g.*, in organic acids.

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An apt survey of the correlations available for calculating the hold-ups in water and in organic media has been given in the book by Nagata⁴ and also in the paper by Vlček and coworkers¹¹ and elsewhere. Here we present the correlation by Calderbank¹ as the one which is best known and most suitable. It is of interest to note that, even though Calderbank has also studied the behaviour of gas dispersions in electrolyte solutions (NaCl, Na₂SO₄, Na₃PO₄), his paper¹ gives just the relationships for calculating the bubble diameter for these solutions but does not mention any for hold-up calculation. The equation given for the bubble diameter in the above solutions of electrolytes significantly differs from those for aqueous solutions of alcohols.

Studies of the rate of coalescence, bubble diameter, hold-up, and foam height and stability make it clear that, almost exclusively, it is the properties of the interface or of thin films which the liquids are capable of producing in-between individual bubbles that are of importance for these processes. The observed effects of salt concentration changes on these quantities in the case of aqueous solutions cannot be accounted for by changes in the commonly studied properties of liquid, *i.e.*, density, viscosity, and surface tension. It has been demonstrated in a number of papers^{1-3,5,6} that small additions of salt to pure water exert a surprisingly strong effect on the bubble coalescence rate. In pure water the bubbles coalesce immediately and large bubbles are formed, producing no foam on the surface. In salt solutions the process of coalescence is retarded, the bubbles are smaller, and foam is produced on the surface. It has been shown that the presence of salts in water also has a strong effect on the properties of the plane phase boundary between gas and liquid⁷.

Hold-up in mechanically agitated solutions of inorganic salts has been studied by Lee and Meyrick⁸. Their analysis yielded an expression for the excess force acting upon the coalescing bubbles in electrolyte solutions. In the agitated tank this force can affect the hold-up value given by a dynamic equilibrium between two mutually opposing phenomena: gas dispersion and bubble coalescence. If two bubbles are to coalesce under the influence of dynamic forces then, also according to Marrucci⁹, they must come into the close proximity to each other so as to form a thin film of liquid in the area of contact. The film thickness is further reduced by stretching. On attaining a critical thickness the film will be ruptured and the bubbles will coalesce. For the case of aqueous electrolyte solutions Lee and Meyrick⁸ derived a relationship for the surface tension gradient:

$$\Delta\sigma = 2/(RT) \cdot c(d\sigma/dc)^2 \cdot \phi, \quad (1)$$

where

$$\phi = 1/[1 + d \ln f/d \ln c] \quad (2)$$

and where σ is the surface tension, R is the universal gas constant, T is absolute temperature, c concentration and f is the activity coefficient. This surface tension gradient prevents the film between the two bubbles or between a bubble and the plane interface from stretching.

The data by Lee and Meyrick⁸ indicate that the hold-up unambiguously tends to rise with increasing value of the quantity $c(d\sigma/dc)^2 \phi$ which we shall denote further by the symbol ψ . Their experimental hold-up data for two salt solutions, *i.e.* of Na₂SO₄ and of NaCl, followed roughly the same lines plotted against the parameter ψ . For these relationships, stirrer rpm and air flow rate were parameters.

Similarly, Meijboom and Vogtländer¹⁰ who have studied the effect of surfactants on the mass transfer from bubbles into a liquid state that the quantity $c(d\sigma/dc)^2$ is well suited for characterizing the drop of surface rate at the liquid-gas interface due to surfactants.

EXPERIMENTAL

Equipment. The hold-up measurements were made using apparatus and procedure described in detail in an earlier paper¹¹. The definition of hold-up follows from the method of determination; the increase in elevation of the surface level due to aeration under the influence of a stirrer is measured against that elevation which results when the stirrer is not activated and no gas is being introduced into the tank. The gas hold-up Z is determined from the level rise data by the formula

$$Z = \Delta\bar{h}/(\Delta\bar{h} + H), \quad (3)$$

where H is the surface level elevation of the liquid in the tank at rest and $\Delta\bar{h}$ is the time-averaged and tank cross section averaged level rise value. The level rise was measured in a cylindrical tank made of organic glass and having an interior diameter $D = 0.29$ m. The elevation at rest, H , was equal to the tank diameter. The tank was fitted with four symmetrically positioned baffles 0.35 m high and 0.03 m wide. At the tank center there was a gas inlet tube of 0.004 m interior diameter. The tube orifice was at the distance of 0.1 m from the tank base. The stirrer used was a six-blade turbine of diameter $d = 0.075$ m mounted at a height of 0.105 m above the tank base.

Air was introduced into the tank from a central distribution main and was led *via* a manostat for pressure equalization, a solenoid valve, and bubblers with fritted discs filled with distilled water and placed in a temperature controller. Further the air passed through a jar filled with glass wool for cleaning to a reducing valve for regulation and to rotameters. The measuring method, worked out in detail earlier¹¹, is based on determining the gas hold-up from the averaged experimental values of liquid level rise due to aeration under the influence of the agitator mounted in the tank. The principle used is to close an electrical circuit by a point electrode in a conductive medium. Two platinum electrodes were used in these measurements; one was a platinum plate of $5 \cdot 10^{-5}$ m² surface area, fixed in a position in the batch near the tank base. The other electrode was fitted so as to permit vertical adjustment. The point electrode was constituted by a platinum wire of $6 \cdot 10^{-4}$ m in diameter. On immersion in the conductive solution studied the electrical measuring circuit was closed permitting passage of electric current. The time of immersion of the movable electrode was indicated by the number of pulses recorded by an electromagnetic counter. The surface level rise of the liquid in tank is a typically local quantity depending on the position of the point electrode with respect to the surface level. It has already been proven¹¹ that the number and position of the measuring points needed to determine average level rise to a sufficient accuracy depend on the type agitator used. With a standard six-blade turbine such as that used in this work it will suffice to limit the measurements to five points along the axis of the sector bounded by two baffles. The individual points were selected so as to characterize equal areas having the form of sectors of an annulus. The radii of the circles on which the measuring points were located were 0.1375, 0.1210, 0.1025, 0.0795 and 0.0460 m.

Before starting the run the value H was read off at the measuring point by contacting the point electrode with the surface level at rest. Two readings were taken at each point during the run. One reading was taken with the electrode adjusted in the position Δh_1 where the time of electrode immersion was longer than half the time interval measured, which was 180 s for all the runs. The other reading was taken with the electrode in a position Δh_2 such that the electrode immersion time was shorter than half the time interval measured. Assuming that these two immersion time values are near to one half of the measured interval — and this assumption was met in our experiments — it is possible to obtain, by line interpolation, the mean value of surface level rise $\Delta\bar{h}$ at the given measuring point. The resultant level rise value $\Delta\bar{h}$, averaged with respect to tank cross section, is an arithmetic mean of five Δh values. The accuracy of the level rise value thus determined is 10^{-4} m¹¹.

The experimental conditions. The measurements were made in solutions of inorganic salts in distilled water. Sodium sulfate solutions of concentrations 0.5 and 1M, magnesium chloride solutions of concentrations 0.5, 1.5 and 2.5M and sodium thiocyanide of concentrations 1, 3 and 5M were used. The hold-up was also measured with distilled water an addition of c. 0.2 g NaCl per liter to increase conductivity. The physical properties of the solutions used are listed in Table I. Most of these were found in tables¹²; the surface tension data for NaSCN were taken from a paper by Jarvis and Scheiman¹³ and NaSCN viscosity was measured in a Höppler viscosimeter. All the measurements were taken at $20 \pm 0.5^\circ\text{C}$. The air flow rates were 7, 14 and 21 liters per min. The stirrer rpm during the runs were 350, 450 and 550 min^{-1} .

RESULTS AND DISCUSSION

Preliminary analysis of the experimental data (Figs 1–3) has confirmed that the quantity ψ can be used to describe the hold-up for various of salts, and the aforementioned assumption has also been confirmed. Marrucci and Nicodemo³ have shown that with rising salt concentration in the bubbled column the bubble diameter decreases down to a certain limiting value, equal for all the salts, and is subject to no further changes at even higher salt concentrations. Thus we have presumed that, beyond a certain concentration, there will be no marked increase in hold-up, too. It has been found that, roughly at values beyond $\psi = 3 \cdot 10^{-6} \text{ kg}^2 \text{ kmol}^{-1} \text{ m}^3 \text{ s}^{-4}$, the hold-up

TABLE I

Physical Properties of the Solutions Used at the Temperature 20°C

Solution	c kmol m^{-3}	ρ kg m^{-3}	$\sigma \cdot 10^3$ kg s^{-2}	η m Pa s
H ₂ O	—	998.0	72.800	1.008
Na ₂ SO ₄	0.5	1 059.5	74.180	1.238
	1.0	1 118.0	75.570	1.576
NaSCN	1.0	1 042.0	72.620	1.161
	3.0	1 117.0	74.000	1.418
	5.0	1 193.0	75.370	1.486
MgCl ₂	0.5	1 036.0	74.310	1.060
	1.5	1 106.0	78.000	1.690
	2.5	1 175.0	81.720	2.660
K ₂ CO ₃	0.04	1 007.0	72.1	1.113
	0.18	1 017.0	72.8	1.141
	0.38	1 032.0	73.5	1.164
	0.74	1 073.0	74.8	1.298

is practically independent of this quantity. This limiting value of the quantity ψ we have denoted ψ_{crit} ; the hold-up can be considered to depend on the dimensionless ratio ψ/ψ_{crit} which we shall denote ψ^+ .

Subsequently, we have attempted to account for the effect of all the quantities important for hold-up in a single equation. Implementing the dimensionless analysis to the fundamental motion equations and the motion equations for two-phase systems in order to describe the behaviour of agitated systems containing a gas phase and a liquid phase we can derive the determining dimensionless criteria of the form¹⁴: $\text{Re} = nd^2\rho/\eta$, $\text{Eu} = P/(n^3d^5\rho)$, $\text{Fr} = n^2d/g$, $\text{We} = n^2d^3\rho/\sigma$, $\text{Kp} = V_g/(nd^3)$.

One of the above criteria, usually the Euler number, is considered to be dependent and is applied to calculate the input. Froude number is used to describe the processes taking place in vessels having no baffles. All our measurements were taken at $\text{Re} > 1.5 \cdot 10^4$ corresponding to turbulent flow during agitation¹⁴. Hence, one could expect the hold-up to be independent of the Reynolds number. The validity of this assumption for our set of data has been tested by Hadačová¹⁵ showing that the effect of the Reynolds number on hold-up was insignificant. In contrast, however, we have assumed on the basis of earlier results that for systems with dissolved salts the above criteria will fall short of yielding an adequate description, inasmuch as the

TABLE I

(Continued)

$\frac{d\sigma/dc}{\text{mol}^{-1} \text{kg m}^3 \text{s}^{-2}}$	ϕ	$\frac{\psi \cdot 10^6}{\text{kmol}^{-1} \text{kg}^{-2} \text{m}^3 \text{s}^{-4}}$	ψ^+
—	—	0.0	0.0
2.769	1.575	6.038	2.010
2.769	1.575	12.076	4.020
0.688	1.000	0.473	0.158
0.688	0.733	1.040	0.347
0.688	0.647	1.530	0.510
3.720	0.905	6.269	2.090
3.720	0.546	11.233	3.780
3.720	0.411	14.225	4.740
3.670	1.000	0.538	0.179
3.670	1.000	2.558	0.852
3.670	1.000	5.110	1.704
3.670	1.000	10.620	3.540

addition of a salt brings about considerable changes in the properties of the dispersion and, thus, also in gas hold-up even though the value of these criteria usually do not change substantially. This is why we have processed the experimental data so as to obtain a regression relationship:

$$Z = Z(We, Kp, \psi^+) . \quad (4)$$

Regression in the usual power form

$$Z = A We^{\alpha_1} Kp^{\alpha_2} \psi^{+\alpha_3} \quad (5)$$

is not suitable, however, since the value of ψ^+ for pure water is equal to zero, whereas the hold-up has a nonzero value. The course of the relationship illustrated in Figs 1–3 suggests that the variable taking account of the quantity ψ^+ should be written in the form $K - \exp(-\psi^+)$. For pure water this variable should have the value of 1, i.e., it should hold that

$$1 = K - \exp(-\psi^+), \quad \psi^+ = 0 \quad (6)$$

yielding $K = 2$. Hence, the resultant regression relationship proposed was

$$Z = B We^{\beta_1} Kp^{\beta_2} [2 - \exp(-\psi^+)]^{\beta_3} . \quad (7)$$

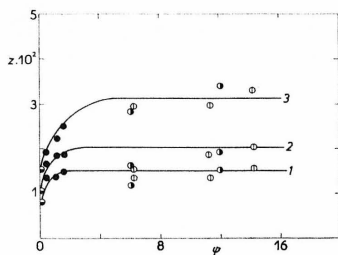


FIG. 1

Gas Hold-up as a Function of the Parameter ψ

$\dot{V}_g = 71 \text{ min}^{-1}$, 1 350 rpm, 2 450 rpm, 3 550 rpm; ● NaSCN, ○ MgCl₂, ● Na₂SO₄, ○ H₂O.

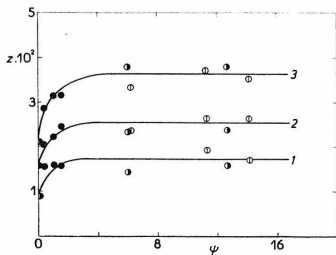


FIG. 2

Gas Hold-up as a Function of the Parameter ψ

$\dot{V}_g = 141 \text{ min}^{-1}$, 1 350 rpm, 2 450 rpm, 3 550 rpm; ● NaSCN, ○ MgCl₂, ● Na₂SO₄, ○ H₂O.

The variable $[2 - \exp(\psi^+)]$ will be denoted further by \bar{Y} . All the experimental hold-up values for the electrolyte solutions as well as for pure water were treated by Eq. (7). Nonlinear regression was used to determine the values of the individual power exponents and of the constant B . The relationship found was

$$Z = 1.3 \cdot 10^{-4} We^{1.00} Kp^{0.36} \bar{Y}^{0.56} \quad (8)$$

The estimates of the constant B and of the variance of the dependent variable Z were $9.4 \cdot 10^{-6}$ and $7 \cdot 10^{-4}$, respectively. The variances of the power exponents β_1 , β_2 , β_3 were estimated as $1.62 \cdot 10^{-3}$, $0.79 \cdot 10^{-3}$ and $2.7 \cdot 10^{-3}$ respectively. The maximum deviation of the calculated hold-up value from the experimental value was 32.5%. The regression function is compared with the experimental data in Fig. 4. Clearly, the type of regression relationship proposed, *i.e.*, Eq. (7), allows to calculate to a sufficient accuracy the values of gas hold-up in solutions of various inorganic salts and in pure water for the given geometrical arrangement. It has been confirmed that the quantity \bar{Y} is adequate for describing the effect of the presence of an electrolyte.

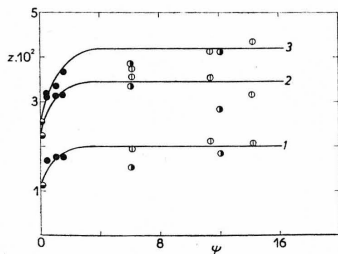


FIG. 3

Gas Hold-up as a Function of the Parameter ψ

$\dot{V}_g = 211 \text{ min}^{-1}$, 1 350 rpm, 2 450 rpm, 3; 550 rpm ● NaSCN, ○ MgCl₂, ○ Na₂SO₄, ○ H₂O.

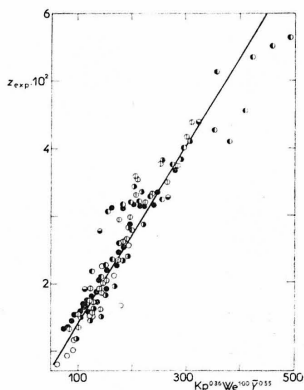


FIG. 4

Comparison of the Relationship $Z = 1.3 \cdot 10^{-4} We^{1.00} Kp^{0.36} \bar{Y}^{0.56}$ with our Data

● NaSCN, ○ MgCl₂, ○ Na₂SO₄, ○ H₂O, ○ K₂CO₃ (ref.¹⁵).

It ought to be mentioned that when numerical values were being substituted into Eq. (8) the value of ψ_{crit} used was equal for all the salts and all the conditions: $\psi_{\text{crit}} = 3 \cdot 10^{-6} \text{ kg}^2 \text{ kmol}^{-1} \text{ m}^{-3} \text{ s}^{-4}$. This value has been estimated from the Figs 1–3 and is open to discussion. The figures indicate that the rise of hold-up increasing concentration fades away at about this value of ψ . More measurements at about this value of ψ would have to be carried out with various kinds of salts to arrive at a precise value of ψ_{crit} .

Comparison with Published Data

In our earlier work¹⁶, hold-up was measured in an apparatus of identical dimensions and of similar geometry but with a considerably larger stirrer having 0.1 m in diameter. Hold-up in water and in K_2CO_3 solutions of four different concentrations (0.04, 0.18, 0.38 and 0.79M) was determined at air flow rates of 5, 15 and 25 liter per min and at stirrer rpm of 300 and 600 min^{-1} . It has been found that the coefficients of Eq. (8) did not fit into these data at higher hold-up values (Fig. 4). Therefore we processed these data independently by nonlinear regression yielding

$$Z = 9 \cdot 10^{-4} \text{ We}^{0.73} \text{ Kp}^{0.36} \bar{Y}^{0.34} . \quad (9)$$

In calculating the quantity \bar{Y} the value of ψ_{crit} used was $3 \cdot 10^{-6} \text{ kg}^2 \text{ kmol}^{-1} \text{ m}^3 \text{ s}^{-4}$. The data treated according to this equation are shown in Fig. 5. It can be seen that even in this case the experimental hold-up values agree well enough with the equation proposed. However, the power exponents for the We number and for the term \bar{Y} differ from those in Eq. (8). This may suggest that their value depends on the geometrical arrangement. We have also processed the data by Lee and Meyrick⁸ using Eq. (7). The experimental values were read off from the figures as the paper gave no data tables. As it has already been said, their hold-up values increased with increasing ψ value. The region of hold-up independence of concentration and thus of the ψ value has not been attained inasmuch as the measurements were performed at lower concentrations, *i.e.*, at values of $\psi < 3 \cdot 10^{-6} \text{ kg}^2 \text{ kmol}^{-1} \text{ m}^3 \text{ s}^{-4}$. Hold-up was measured in NaCl and Na_2SO_4 solutions at four stirrer rpm levels and three gas flow rates. Correlation of their data by Eq. (7) gave

$$Z = 5.38 \cdot 10^{-4} \text{ We}^{0.84} \text{ Kp}^{0.56} \bar{Y}^{1.41} . \quad (10)$$

A comparison of the experimental data with this equation is shown in Fig. 6. The data points for pure water were not included. Even though the hold-up values as determined by Lee and Meyrick⁸ for the highest concentrations used were by up to 200% higher than those for pure water, and the differences in hold-up were up to 150% between 300 and 600 rpm at the highest concentrations, the authors found that

the rpm dependence of hold-up in pure water was very slight. For instance, at the gas flow rate of 16 liter per min the data are practically independent of rpm. This, of course, contradicts the findings of a number of authors, *e.g.* of Nagata⁴ and it also is at variance with our results, see Figs 1–3. Lee and Meyrick⁸ took their measurements using an apparatus similar in geometry and size to that of Dolejš¹⁶. They used a sixblade turbine stirrer of cca 0.1 m in diameter. Therefore the difference encountered between the respective coefficient of Eqs (9) and (10) is surprising. The only difference between the two apparatuses was the air inlet: in that by Dolejš¹⁶ the air inlet was just below the stirrer, as it also was in our experiments, and the inlet tube orifice had 4 mm in diameter, whereas in that by Lee and Meyrick⁸ the air was introduced through an orifice in the tank base having 1.6 mm in diameter.

Trace amounts of surface-active contaminants having a significant effect on the surface properties of liquids might constitute still another source if incompatibility between experimental data presented by different authors.

As shown in the paper Vlček and coworkers¹¹ the method of hold-up measurement suggested by Lee and Meyrick⁸ is less accurate than is our method. This is also testified to by the erroneous data for water. Hence, the only conclusion that can be drawn

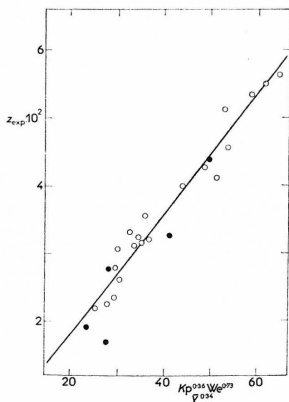


FIG. 5

Comparison of the Relationship $Z = 9 \cdot 10^{-4} We^{0.73} Kp^{0.36} \bar{\gamma}^{0.34}$ with the Data by Dolejš (ref.¹⁵)

○ K_2CO_3 , ● H_2O .

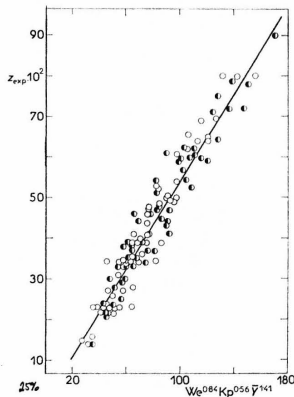


FIG. 6

Comparison of the Relationship $Z = 5.38 \cdot 10^{-4} We^{0.84} Kp^{0.56} \bar{\gamma}^{1.41}$ with the Data by Lee and Meyrick⁸

○ NaCl, ● Na_2SO_4 .

on the basis of processing their data is that the type of regression is adequate but the parameter values can apparently be doubted.

It ought to be mentioned that Dolejš¹⁶ used the solution of just one salt, so that the value of ψ was only dependent on concentration; the parameter $d\sigma/dc$ was not being changed.

CONCLUSIONS

The paper contributed to finding suitable calculational relationship that would adequately express the gas hold-up in liquid batches aerated under the influence of agitation. The effect of the presence of inorganic salts dissolved in water on the hold-up value was studied and the results were compared with experimental data presented by other authors. The results can be summarized as follows: 1) It has been confirmed that for the purpose of comparing hold-up data for solutions of different salts the quantity $\psi = c(d\sigma/dc)^2\phi$ can be recommended. 2) In electrolyte solutions the hold-up increases with increasing ψ at otherwise equal conditions, up to a certain value of this parameter estimated as $\psi_{crit} = 3 \cdot 10^{-6} \text{ kg}^2 \text{ kmol}^{-1} \text{ m}^3 \text{ s}^{-4}$. Beyond this value the hold-up remains practically constant at given rpm. 3) The hold-up can be adequately described quantitatively by the relationship $Z = B \text{ We}^{\beta_1} \text{ Kp}^{\beta_2} \bar{Y}^{\beta_3}$, where $\bar{Y} = 2 - \exp(-\psi/\psi_{crit})$. 4) For the geometrical configuration of our apparatus and for the salt solutions used in this work the equation assumes the form

$$Z = 1.3 \cdot 10^{-4} \text{ We}^{1.00} \text{ Kp}^{0.36} \bar{Y}^{0.56} .$$

For other geometries the power exponents may be different.

LIST OF SYMBOLS

a	activity, N L^{-3}
c	concentration, N L^{-3}
D	interior diameter of tank, L
d	stirrer diameter, L
f	activity coefficient
g	acceleration due to gravity, L T^{-2}
H	surface level elevation in tank at rest, L
$\Delta\bar{h}$	time-averaged local level rise, L
$\Delta\bar{h}$	level rise averaged with respect to tank cross section, L
n	stirrer rpm, T^{-1}
P	stirrer input, $\text{M L}^2 \text{T}^{-3}$
R	gas constant, $\text{M N}^{-1} \text{L}^2 \text{T}^{-2} \theta^{-1}$
T	absolute temperature, θ
V_g	gas flow rate, $\text{L}^3 \text{T}^{-1}$
$\bar{Y} = 2 - \exp(-\psi^+)$	
Z	hold-up, defined by Eq. (3)

$\alpha_1, \alpha_2, \alpha_3,$	constants
$\beta_1, \beta_2, \beta_3,$	constants
ϕ	correction factor defined by Eq. (2)
ψ	coalescence rate characteristic, $M^2 N^{-1} L^3 T^{-4}$
ψ_{crit}	critical value, $M^2 N^{-1} L^3 T^{-4}$
ψ^+	dimensionless quantity
η	dynamic viscosity of batch, $ML^{-1} T^{-1}$
ρ	batch density, ML^{-3}
σ	surface tension of batch, MT^{-2}

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