

Residence time distribution of the gas phase in a mechanically agitated gas-liquid reactor.

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ABSTRACT

In this study we present a measuring method and extensive experimental data on the gas phase RTD in a mechanically agitated gas-liquid reactor with standard dimensions over a wide range of superficial gas velocities, agitation rates and agitator sizes. The results are modelled successfully, using the weighed moments method, by a simple RTD model of one mixer in series with a plug flow zone. All results are correlated in one relation, which can be used for scale-up. As a side rule, the following relation for the gas hold-up has been obtained:

$$\epsilon_g = \frac{(0.88 * u_g + 5.1 * 10^{-3})}{H} + 3.09 * 10^{-2} * \frac{(n - n_0) * (\frac{D_i}{D})^2}{H}$$

with $h_r/D = 0.33$, $n \geq 1.5 * n_0$, $D = H$, $H = 0.286$ m, $0.3 \leq D_i/D \leq 0.5$,
 $4.9 * 10^{-3} \leq u_g \leq 3.03 * 10^{-2}$ m/s

The correlation error is within 20%, and is in good agreement with the holdup correlation derived by Van Dierendonck (1970).

KEYWORDS

Residence time distribution, gas holdup, gas-liquid reactor, modelling, scale-up.

INTRODUCTION

The residence time distribution RTD of the gas phase in a mechanically agitated gas-liquid reactor is not very well known, see e.g. Shah (1979). The RTD of the gas phase determines the driving force for mass transfer between the gas and liquid phase and thus is of direct importance for the yields and selectivities in gas-liquid reactors. Especially if the conversion of the gas phase reactant is high, the influence of the gas phase RTD on the reactor performance can be pronounced. Knowledge of the gas phase RTD is also important for the experimental determination of gas-liquid mass transfer parameters, particularly in case of high degree of gas phase conversions for gas phase reactants or high absorption fractions of key components.

The purpose of our work was to elaborate a reliable basis for prediction of RTD, allowing a scale up for G/L systems. In this paper we present extensive experimental data on the gas phase RTD in a mechanically agitated gas-liquid reactor over a range of superficial gas velocities, from $4.9 * 10^{-3}$ m/s to $3.03 * 10^{-2}$ m/s. All

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results are correlated in one algebraic expression, which can be used for scale-up and design calculations as well as for mass transfer parameter evaluations.

LITERATURE

Despite the vast amount of research on mechanically agitated gas-liquid reactors, there are only very limited data available on the gas phase RTD. These are mainly qualitative and not really conclusive. Hanhart *et al.* (1963) concluded that, in the region where the agitation is effective ($n > n_0$), the gas phase can be considered ideally mixed. Below the critical agitation rate, n_0 , the gas phase RTD resembled plug flow. Gal-Or and Resnick (1966) found that a cascade of two ideally mixed tank reactors gave the best representation. Chapman *et al.* (1982) observed that for high gas velocities the gas phase RTD more approaches plug flow, while for low gas velocities the gas phase RTD was intermediate between that of an ideally mixed tank reactor and a plug flow reactor.

Van Dierendonck (1970) has measured the gas hold-up in a mechanically agitated gas-liquid reactor, by determining the height difference between the dispersed and the non-dispersed liquid. Measurements were carried in mechanically agitated gas-liquid reactors with different vessel diameters and stirrer diameters. Van Dierendonck correlated his experimental data:

$$\epsilon_g = 0.31 * \left(\frac{u_g}{\sqrt{\left(\frac{\sigma * g}{\rho} \right)}} \right)^{2/3} + 0.45 * \frac{(n - n_0) * \left(\frac{D_i^2}{D} \right) * \left(\frac{D}{H} \right)}{\sqrt{(g * D)}} \quad (1)$$

$$\text{with } \epsilon_g \leq 0.25, \frac{(n - n_0) * \left(\frac{D_i^2}{D} \right)}{\sqrt{(g * D)}} < 0.5, \quad \frac{h_R}{D} = 0.5, \\ n \geq n_0, 0 \leq u_g \leq 0.05 \text{ m/s}, 0.7 < H/D < 1.4, D < 3 \text{ m}$$

The critical rate n_0 can be predicted by literature equations of Westerterp (1962) and of Van Dierendonck (1970). The critical agitation rate is defined as the agitation rate by which the extension of the straight line, which represents the relation between ϵ_g and n , intersects the n -axis. The relations show both a dependency of the reaction system geometry and the physical properties of the liquid. The equation of Van Dierendonck is not representative, because the equation is defined for $h_R/D = 0.5$. In our reactor $h_R/D = 0.66$, so the critical agitation rates can only be used as an indication. For the three impellers used, the critical agitation rates are determined with the relation given by Westerterp, and are given in Table 1.

Table 1: Critical agitation rates

D_i/D (-)	Westerterp n_0 (rps)	Van Dierendonck n_0 (rps)
0.3	10.2	10.3
0.4	6.2	5.8
0.5	4.2	3.7

THEORY

Evaluation of the measurements by the weighed moments method

We selected the negative step disturbance measuring principle. An experimental, negative step disturbance results in a Wash-out diagram., the so called W-diagram. With the W-diagram it is possible to calculate the

distribution diagram. The distribution function $E(t)$ gives the chance $E(t)dt$ of a volume element to have a residence time between t and $t+dt$ seconds. $W(t)$ can be defined as the chance of volume element to have a residence time longer than t . To characterize $E(t)$ in terms of statistical parameters, like average residence time and the spread in residence time, the moments method is used. The n th moment is defined by

$$M_n = \int_0^{\infty} t^n * E(t)dt = n * \int_0^{\infty} t^{n-1} * W(t)dt \text{ with } W(t) = 1 - \int_0^t E(t)dt \quad (2)$$

The average residence time and the variance can easily be determined from the first and second moment. Westerterp et al (1973) describe the introduction of the Laplace factor e^{-st} , which diminishes the influence of disturbances in the tail. Weighed moments are then obtained.

Two independent methods, using weighed moments, can be used to determine the best fitting model. The methods are only suitable for simple models. The first method calculates directly process parameters for a model from the weighed moments, using Laplace domain analysis. The second method developed by Michelsen et al (1970) calculates the model process parameters by linear relations between the weighed moments and the Laplace variable s . Both methods have been applied to some of our experimental results, we found no significant difference in the calculated parameters. Because the first method takes much more computer time, due to the use of a convolution integral, the method of Michelsen et al has been selected by us. The Michelsen method derives a relation between the weighed moments and the Laplace variable s . Hopkins et al (1969) recommended a value for the dimensionless group $s\tau$ in the range of: $2 \leq s\tau \leq 5$. Three models are selected to be used to interpret the experimental results; model (1), the cascade of equally sized, ideally mixed tanks, model (2), the cascade of equally sized, ideally mixed tanks in series with a tube with plug flow and model (3), the single, ideally mixed tank in series with a tube with plug flow. The distribution functions for these models are known from literature, see Westerterp et al (1973), or can easily be derived. From these functions the moments, the Laplace transformed distribution functions and the weighed moments can be calculated, as is shown by Pham and Key (1977). The relations derived for the weighed moments can be rewritten into the linear relations between the moments and s , analogous to Pham and Key.

Influence of tracer absorption

Joseph et al (1986) have examined the effect of the solubility of the tracer gas on the accuracy of the dispersion measurement, using the axial dispersion model with interphase transfer. For the case of a batch liquid and the gas phase being completely back mixed the following equation for the first moment was derived

$$M_1 = 1 + \frac{e_L}{m * e_g} \quad (3)$$

For the second moment the error is even bigger, compared to the first moment. The least soluble gas in water at room temperature is Helium ($m = 106$). Corrections for tracer solubility should be done by using time domain analysis to compare measured and predicted RTD curves rather than the method of moments.

EXPERIMENTAL

Installation.

The flow sheet of the installation is given in Fig. 1. The reactor is equipped with an inverted collecting funnel, by which a part of the gas from the reactor is sucked off. The installed impeller can readily be exchanged by another one, so that the influence of the impeller size can be studied also.

The reactor. The mechanically stirred reactor, has a diameter of $D = 286$ mm and a total height equal to 400 mm. The reactor has four vertical baffles each of width $W/D = 0.1$, and is, inclusive the baffles, made of transparent perspex. The reactor operates continuously with respect to both the gas and the liquid phase. Three six-bladed disc turbine impellers are used, with diameters, D_i , of respectively 0.3, 0.4 and 0.5 times the reactor diameter. The stirrer is installed at a height of $h_R/D = 0.33$ above the flat bottom plate. The dispersion level during all experiments, is maintained at a height of $H/D = 1$ by adjusting the amount of

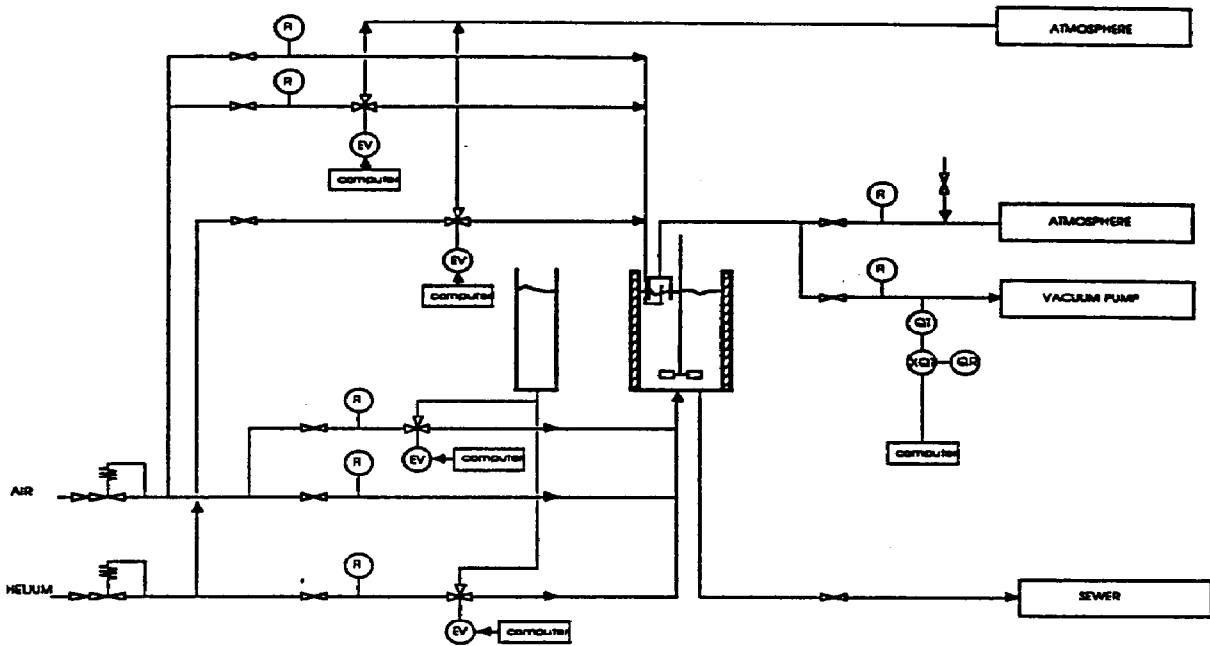


Figure 1: Experimental installation

liquid. A gas mixture of helium and air is introduced into the reactor via a sparger located centrally below the impeller, the sparger being a simple tube with a diameter of $d_s = 3.0$ mm. The agitation rate is adjusted with a variable gear motor and is monitored continuously with a pulse-counter.

The collecting funnel. The funnel, as shown in Fig. 2, serves as inlet to the detection system, through which a part of the gas from the dispersion is sucked off, via a vacuum pump at 300 mm Hg. The construction of the funnel is critical, since it has to be as small as possible to minimize the dead volumina in the detection system, and must have such a shape that no liquid can enter into the system despite the fluctuations of the dispersion level. These two demands led to the final shape with a sloping wall and breakwaters. It is located in the dispersion in such a way that no gas from outside the reactor is sucked into the funnel.

To obtain the true RTD of the reactor, it is necessary to account for the RTD of the detection system consisting of the funnel and a catharometer. To this end, the latter is measured by introducing into the funnel a gas mixture of helium and air directly via a blowpipe, located centrally below the funnel and just at the level of the dispersion surface.

The gas system. Helium is taken from a gas cylinder while air is supplied by a compressor. After pressure adjustment the gases are supplied to the reactor and to the funnel system, each via three Brooks Rotameters. The gas flow to the reactor and the funnel consists of a main air stream and a auxiliary tracer gas stream, the latter containing either pure helium as tracer gas or air. The switching between helium and air is carried out with two electronic magnetic valves. The flow rate of the tracer gas streams range from $0-0.16 \cdot 10^{-3}$ m^3/s and from $0-1.6 \cdot 10^{-6}$ m^3/s for the reactor and blowpipe, respectively. The flow rate for the main and carrier air streams range from $0-3.5 \cdot 10^{-3}$ m^3/s and $0-32 \cdot 10^{-6}$ m^3/s for the reactor and the funnel, respectively. The gas in the funnel is led to a Thermal Conductivity Detector, which measures the helium concentration. Distilled water is used as liquid.

Procedure.

The experiment. All rates and flows are set to the desired values. About 95% of the gas flow supplied in the funnel from the dispersion and the blowpipe together is sucked off, to avoid entraining of liquid into the catharometer. Then the dispersion level is adjusted to a height of 286 mm, followed by lowering the funnel and blowpipe just to the level of the dispersion surface. As an example the helium concentration in the reactor inlet is 5 vol % and in the funnel zero. At $t=0$ a step disturbance is created by switching from helium to air. A sharp and a well reproducible step is created, by using electronic magnetic valves. The computer controls the experiments; two series of 10 experiments are carried out without interruption. That is a series of 10 RTD experiments for the entire system of

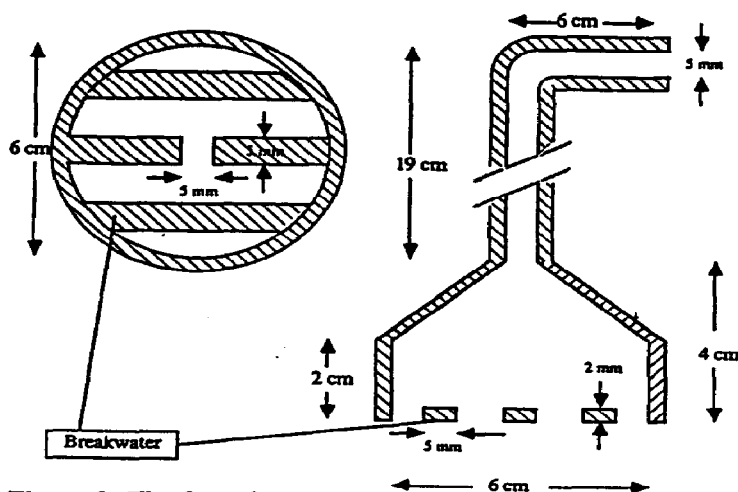


Figure 2: The funnel

reactor and detection system, followed by 10 RTD experiments for the detection system only. To evaluate the experiments, the first and second moment of the two series of 10 RTD measurements are calculated in order to obtain the average residence time and the variance. The measurements are averaged, after which the first and second moment of the averaged measurements for the whole system and for the detection system are calculated for 10 values of s , so that the process parameters of the three models can be derived. The moments for the reactor itself can be determined by subtracting the values of the detection system from those for the whole system. This is only allowed if the two RTD measurements are independent. This is true if the relative variance in the detection system is constant for all experimental conditions. Experimental results showed no influence of the stirrer diameter or agitation rate, so it is reasonable to assume the two systems to be independent.

Funnel position and dead volumina. By using a funnel collecting only part of the gas from the dispersion, the possibility of radial or tangential differences had to be checked on experimentally. Several experiments were done under the same conditions but with different locations of the funnel; near the wall, directly behind a baffle, and in the middle of a quadrant. From the results, no significant difference could be detected. For further systematic experiments the location in the centre of the vessel is selected, because there the fluctuations of the surface are lowest.

RESULTS

Experimental conditions.

Totally 85 experiments are carried out at atmospheric pressure and normal temperature (± 20 °C), by which three process parameters are varied. Measured is at four gas velocities, three impeller diameters and various agitation rates. The superficial gas velocity was chosen in the range of $4.9 \cdot 10^{-3}$ to $3.03 \cdot 10^{-2}$ m/s. The impeller diameters were 0.3, 0.4 and 0.5 times the reactor diameter. The agitation rate varied for each gas velocity and impeller diameter from about one below to about five above the critical agitation rate, ranging from 4.6 to 25 rps.

Experimental results

The experimental measured residence times are shown in Table 2. From the experimental results and the Michelsen method the best fitting model was determined. This is done by determining the correlation coefficients of the linear relations by the Michelsen method. Results show the best fit with model (2), almost the same results were found with model (3), followed by a rather poor fitting for model (1). The difference in the quality of the fit between model (2) and (3) is small. Model (3) is, in our view, the most realistic

approach, compared to the non-integer number of mixers in the cascade in model (2). In Fig. 3 the number of tanks in the cascade, for model (2), is given for stirrer speeds above $1.5 \cdot u_0$.

Table 2: Experimental results.

D_i/D	$u_s = 4.9 \cdot 10^{-3}$ m/s		$u_s = 1.0 \cdot 10^{-2}$ m/s		$u_s = 1.91 \cdot 10^{-2}$ m/s		$u_s = 3.03 \cdot 10^{-2}$ m/s	
	n (rps)	τ (sec)	n (rps)	τ (sec)	n (rps)	τ (sec)	n (rps)	τ (sec)
0.3	11.7	3.52	12.5	2.57	13.4	1.93	16.7	1.83
0.3	13.8	4.72	14.1	2.83	15	2.05	20.2	2.08
0.3	15	5.59	15.8	3.31	16.7	2.43	21.8	2.10
0.3	16.7	6.30	17.5	3.28	18.3	2.65	23.5	2.33
0.3	19.1	7.20	19.1	3.84	20.1	2.72	25.1	2.29
0.3	20.8	7.78	21.7	4.21	21.9	3.19	26.9	2.41
0.4	6.7	3.67	5.8	2.26	5.8	1.65	5.1	1.10
0.4	8.3	4.89	7.6	2.48	7.1	2.19	6.8	1.44
0.4	10.1	6.58	9.2	3.43	8.4	2.17	8.5	1.69
0.4	11.8	7.54	10	3.90	10.2	2.50	10	2.14
0.4	12.5	7.42	11.7	4.29	11.7	2.94	13.4	2.54
0.4	10.8	6.39	12.6	4.54	13.4	2.94	15.2	2.73
0.5	4.2	3.67	4.6	2.84	5.8	2.54	5	1.62
0.5	5	5.07	5.4	3.22	6.6	2.68	5.9	2.04
0.5	5.8	5.69	6.3	3.88	7.5	2.86	6.7	2.20
0.5	6.6	6.03	7.1	3.95	8.3	3.23	7.6	2.31
0.5	7.5	7.52	7.9	4.95	9.3	3.51	8.4	2.41
0.5	8.3	8.62	8.8	5.56	10	3.79	9.2	2.43

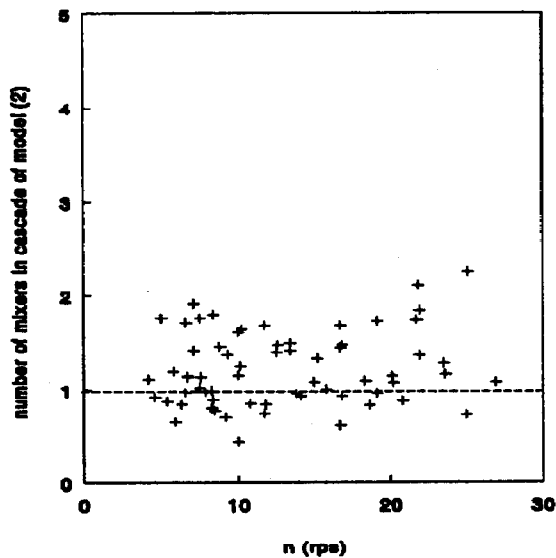


Figure 3: Number of mixers in cascade of model (2)

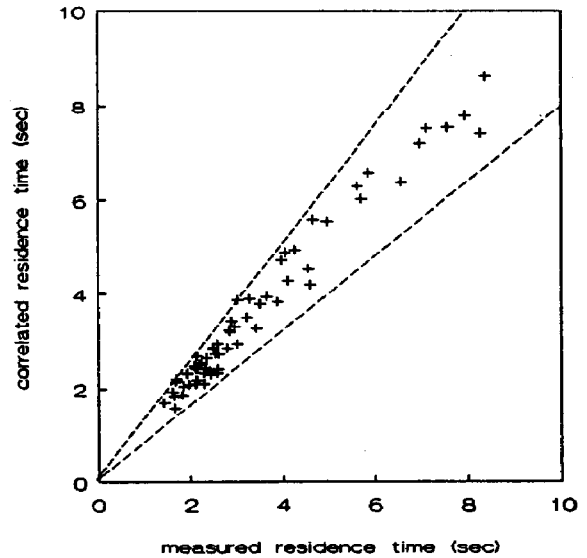


Figure 4: Parity plot

Proceeding from the most realistic model, model (3), it is attempted to correlate the experimental liquid holdup. First the rotational speed dependency, on the residence time of the single mixer is determined. For

each gas velocity and stirrer diameter a relation is derived, showing a linear dependency of the stirrer speed minus the critical stirrer speed; $(n - n_0)$. For the gas velocity an inversely proportional, and for the stirrer vessel diameter ratio a square dependency was found. The residence time found is correlated by:

$$\tau_{\text{mixer}} = 3.09 \cdot 10^{-2} \cdot \frac{(n - n_0)}{u_g} \cdot \left(\frac{D_i}{D}\right)^2 \quad (4)$$

The model verification results showed a constant residence time in the plug flow tube at constant gas velocity. The dependency of the gas velocity on the residence time in the plug flow section is inversely proportional, resulting in:

$$\tau_{\text{plug}} = 0.88 + \frac{0.51 \cdot 10^{-2}}{u_g} \quad (5)$$

The overall residence time is given by:

$$\tau = \tau_{\text{mixer}} + \tau_{\text{plug}} \quad (6)$$

with $n > 1.5 \cdot n_0$ and $D = 0.286$ m

A parity plot is created for the model residence time versus the experimental residence time, as shown in Fig. 4. The error is within 20 % for $n > 1.5 \cdot n_0$. The residence time relation can be rewritten to the gas holdup relation:

$$e_g = 3.09 \cdot 10^{-2} \cdot \frac{(n - n_0)}{H} \cdot \left(\frac{D_i}{D}\right)^2 + 0.88 \cdot \frac{u_g}{H} + \frac{5.1 \cdot 10^{-3}}{H} \quad (7)$$

with $h_R/D = 0.33$, $n \geq 1.5 \cdot n_0$, $D = H$, $H = 0.286$ m, $0.3 \leq D_i/D \leq 0.5$, $4.9 \cdot 10^{-3} \leq u_g \leq 3.03 \cdot 10^{-2}$ m/s

DISCUSSION AND CONCLUSIONS

Tracer absorption may cause errors in the experimental results. Our gas holdup correlation is only suitable for agitation rates higher than $1.5 \cdot n_0$, resulting in a rather high gas holdup. According to the correction by Joseph et al (1986), the error is within 15 %. However, the method directly calculating the process parameters, uses the Laplace domain analysis, giving the same results as the method developed by Michelsen et al (1970). So, tracer absorption apparently did not create errors in the RTD measurements.

Our RTD results can be compared to those of Van Dierendonck (1970). The only difference between our and van Dierendonck's experiments is the h_R/D value. Van Dierendonck proposed a gas holdup contribution, see formula (1), of the agitation rate and a contribution of the gas fraction. Both contributions can directly be compared to our gas holdup relation, assuming the agitation rate contribution to be the mixer part, and the gas fraction contribution to be the plug flow part. The mixer contributions have same dependencies for the agitation rate and the impeller diameter, but have a different constant factor. This has to be attributed to the different h_R/D ratio.

Our gas holdup correlation comprises an agitation rate independent and an agitation rate dependent contribution. This is in agreement with the physical model assumed: single ideally mixed tank in series with a tube with plug flow. At low agitation rates the plug flow part is dominant, describing the RTD of the gas phase by a tube with plug flow. At high agitation rates the mixer part becomes more important, by which at extreme high agitation rates the RTD of the gas phase corresponds to a single ideally mixed tank.

We conclude that the RTD of the gas phase in a mechanically agitated gas-liquid reactor can be described by a single ideally mixed tank in series with a tube with plug flow. The Michelsen method we have used in this study yields virtually the same results as the Laplace domain analysis.

The gas holdup in a mechanically agitated gas-liquid reactor, for the system water-air, can be described by correlation (7). This correlation is useful for scale-up, in combination with the by Van Dierendonck's (1970) found reactor diameter dependency and physical properties dependency.

NOMENCLATURE

d_s	sparger diameter	m	n_0	critical agitation rate	s^{-1}
d_f	funnel diameter	m	t	time	s
D	reactor diameter	m	u_s	superficial gas velocity	m/s
D_i	impeller diameter	m	W	baffle width	m
$E(t)$	distribution function		$W(t)$	wash out function	
g	gravity constant	m/s^2			
h_f	funnel height	m	ϵ_g	gas holdup	-
h_R	stirrer height above bottom	m	ϵ_l	liquid holdup	-
H	dispersion height	m	ρ	density	kg/m^3
m	Henry coefficient	-	σ	surface tension	N/m
M_n	nth moment		τ	residence time	s
n	agitation rate	s^{-1}			

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