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# DIAMETER OF BUBBLES IN BUBBLE COLUMN REACTORS OPERATING WITH ORGANIC LIQUIDS

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In order to determine physically justified relations between bubble characteristics and the physicochemical properties of liquid employed, bubble size distributions and average bubble diameters were calculated using theoretically justified model of gas bubbling through the liquid layer. The model of the gas bubbling enabling the calculations of the bubbles diameter has been based on the original approach by Prince and Blanch. It assumes an equilibrium between the coalescence and dispersion processes, and uses a simplified method of solution of the population balance equations. This model has been verified experimentally for different organic liquids (acetaldehyde, acetone, cyclohexane, isopropanol, methanol, n-heptane, toluene) and columns of different scale: glass laboratory column 9 cm diameter and 200 cm high, operated at atmospheric pressure and low temperature; stainless steel pilot plant column 30.4 cm diameter and 400 cm high, operated at elevated pressure (up to 1.1 MPa) and temperature (up to 160°C). The model has been subsequently used to establish a theoretically based correlation for the bubble diameter by means of a numerical experiment using 'virtual liquids'.

*Keywords: bubble column; bubble diameter; mathematical modelling; numerical experiment; 'virtual liquids'.*

## INTRODUCTION

Many industrially important reactions, such as oxidations, hydrogenations, halogenations, are carried out in gas–liquid reactors, using gas bubbling as the means of contacting of the reacting phases. Usually the reacting liquid is organic, and the reactor is operated at elevated temperature and pressure. A good example of such a process is the CYCLOPOL process (Krzysztoforski *et al.*, 1986), in which cyclohexane is oxidized in the liquid phase to form cyclohexanol and cyclohexanone, later converted to caprolactam. In order to improve the selectivity of the process, an exhaustive research programme was carried out during the last 20 years, successfully completed in 2003 (Pohorecki *et al.*, 2003, 2004). Within this programme a mathematical model of the reaction kinetics was developed for the reactions involved, as well as a model of the reactor hydrodynamics; both models were experimentally verified, and used for the general description of the process (Pohorecki *et al.*, 2001a, b). Eventually the improvements of the existing process have been proposed and successfully implemented in the industrial practice. The hydrodynamics part of the research programme is described in the present paper.

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## HYDRODYNAMICS OF THE BUBBLE COLUMNS

In spite of widespread industrial application of bubble systems, the hydrodynamics description of such systems is far from being complete. There exists a number of models, describing gas–liquid flows at different scales (interface tracking models for single bubble, Euler–Lagrange models for bubble swarms, Euler–Euler models for the whole apparatus). These models can be coupled to give a multi-level model (van Sint Annaland *et al.*, 2003; Deen *et al.*, 2004).

In many cases, however, the bubble diameter has to be assumed a priori, and to this end empirical and semi-empirical correlations are normally used. Such correlations are usually based on a limited number of experiments, and often give highly divergent results when applied to a case at hand.

In particular, different correlations predict very different effect of liquid properties on the bubble size. Table 1 shows the influence of liquid properties on the average bubble diameter, as described by different correlations. As it is seen, the exponent on the liquid density changes in the range from 0 to $-0.74$, that on the liquid viscosity changes in the range 0–0.24, and that on the interfacial tension changes in the range 0.03–0.6. This is caused by the fact that it is impossible to change any of the above properties without changing the other two, and moreover, the range of changes realizable using easily accessible liquids is

*Table 1.* Influence of the liquid properties on the bubble diameter according to existing correlations.

| | Correlation | Liquid density | Liquid viscosity | Surface tension |
|---|---|---|---|---|
| 1 | Hughmark (1967) | $\rho_L^{-0.2}$ | $\mu_L^0$ | $\sigma^{0.6}$ |
| 2 | van Dierendonck (1970) | $\rho_L^{-0.5}$ | $\mu_L^0$ | $\sigma^{0.5}$ |
| 3 | Akita and Yoshida (1974) | $\rho_L^{-0.74}$ | $\mu_L^{0.24}$ | $\sigma^{0.5}$ |
| 4 | Kumar *et al.* (1976) | $\rho_L^{-0.25}$ | $\mu_L^0$ | $\sigma^{0.25}$ |
| 5 | Idogawa *et al.* (1986) | $\rho_L^0$ | $\mu_L^0$ | $\sigma^{0.08\,a}$; $\sigma^{0.03\,b}$ |
| 6 | Idogawa *et al.* (1987) | $\rho_L^{-0}$ | $\mu_L^0$ | $\sigma^{0.20\,a}$; $\sigma^{0.08\,b}$ |
| 7 | Wilkinson (1991) | $\rho_L^{-0.45}$ | $\mu_L^{0.22}$ | $\sigma^{0.34}$ |

[a] $P = 0.1$ MPa; [b] $P = 1.0$ MPa.

very limited. Yet in many cases a prediction of bubble diameter is necessary for a liquid, whose properties are outside the ranges investigated. To overcome this difficulty, and to develop a theoretically based correlation for bubble diameter, a numerical experiment has been used in this work.

In any bubble column four different regions may be distinguished (Millies and Mewes, 1999):

(1) the region of primary bubbles (produced by the gas distributor);
(2) the region of secondary bubbles (produced by break-up of the primary ones);
(3) the region of dynamic equilibrium between coalescence and disruption of the bubbles;
(4) the separation region, at the top of the liquid layer.

In sufficiently deep liquid layers (e.g., in bubble columns), most of the column volume is occupied by the third (coalescence/redispersion) region. In this region the bubble characteristics may be expected to depend on the gas velocity and liquid properties, and not on the geometry of the gas distributor. This region is the subject of the present work

The research programme comprises the following parts:

(1) development of a general, theoretically justified model of gas bubbling through a liquid layer;
(2) verification of the model proposed in a wide ranges of temperature and pressure, using different gas–liquid systems and equipment of different scales;
(3) performing a numerical experiment using the verified model, in order to develop a physically justified correlation, enabling simple calculation of bubble diameter for different liquids.

## MODEL CALCULATIONS

A simplified version of the model developed originally by Prince and Blanch (1990) was used.

The population balance equation used was of the form suggested by Fleisher *et al.* (1996):

$$\frac{\partial}{\partial t}n(z, d_b, t) + \frac{\partial}{\partial z}[n(z, d_b, t)u_r(z, d_b)] + \frac{\partial}{\partial d_b}\left[n(z, d_b, t)\frac{\partial}{\partial t}d_b(z, d_b)\right] = G(z, d_b, t) \qquad (1)$$

where the first term describes the change of bubble number concentration with time, the second is the convection term, the third describes bubble growth, and the right hand side is the generation function. For the equilibrium region considered in this work, we observed experimentally that the bubble size distribution does not change in time or along the column axis. Moreover, in the absence of mass transfer and with sufficiently small pressure change, one can assume that all the terms on the left hand side are equal to zero. Dividing the total bubble population into $N$ classes one can write equation (1) as:

$$G_i = 0 \qquad (2)$$

where $G_i$ is the generation function for bubbles of class 'i'.

The generation function is the difference between bubble birth and death functions. The bubble 'births' in a given class result from breaking a bigger bubble, or from the coalescence of smaller bubbles. Assuming that a bubble can be broken into two smaller bubbles of equal volume (which is rather arbitrary assumption) or be formed by coalescence of two smaller ones, we can write:

$$G_i = \frac{1}{2}\sum_{k=1}^{N}\sum_{l=1}^{N} C_{i,kl} - \sum_{j=1}^{N} C_{ij} + 2B_m - B_i \qquad (3)$$

where

$$v_m = 2v_i \qquad (4)$$

and

$$C_{i,kl} = \begin{cases} C_{kl} & \text{if } v_k + v_l = v_i \\ 0 & \text{if } v_k + v_l \neq v_i \end{cases} \qquad (5)$$

The model assumes that:

(1) the bubble coalescence rate is equal to the product of the bubble collision rate and the collision efficiency;
(2) the bubble collisions may be caused by turbulence, buoyancy or laminar shear;
(3) the bubble break-up rate is equal to the product of collision rate of bubbles and turbulent eddies and collision efficiency;
(4) bubbles are broken by eddies of the same size as the bubble or smaller (but not smaller than 20% of bubble diameter);
(5) the bubble–eddy collision efficiency depends on eddy kinetic energy.

The details of the model used and the results obtained using this model have been described in our two earlier papers (Pohorecki *et al.*, 2001a, b).

In the conditions considered the bubble size distribution can be described by a log-normal distribution. The parameters of the distribution were selected so as to minimize the $\sum_i G_i^2$ values [equation (3)]. Bubble concentration can be obtained from the gas hold-up and the average bubble