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GAS-HOLD-UP AND BUBBLE SIZE DISTRIBUTION IN A FIBROUS FIXED BED BIOFILM REACTOR

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Abstract

In this paper, the gas-liquid dispersion in a biofilm reactor with fibrous structured packing is studied. The reactor is operated with Pseudomonas putida aimed in waste water treatment. Gas hold-up and bubble size distribution are determined. The properties of the gas dispersion proved to be stabilized by the fibrous bed presence and showed improvement of the dispersion parameter by the packing. Apart from showing high gas hold-up and low bubble size due to surface active and viscous dissolved elements, the biochemical reaction did not pose any significant additional effect. The results are shown to be comparable with previous studies of mesh wire packing. An empirical equation is proposed that relates gas hold-up with all studied variables.

Key words: *Fibrous fixed bed biofilm reactor, Gas hold-up, Bubble size distribution*

1. INTRODUCTION

Gas-liquid bubble columns with structured packing have large potential of applications in biochemical processing [1]. Alternatively, a packing itself is attractive for biomass immobilization forming biofilms for large-scale biological treatment. Being known to enhance transversal viscous flow in viscous batches, structured packing is prospective for complex non-Newtonian bio-fluid flow stabilization. At high fluid velocity, they could serve as replacement of highly energy consuming mechanical mixers in mixing bioreactors. The applications are multi-functional and require additional studies to uncover the character of functional variation in the case of the particular internal body. The study is targeted at the first option of low velocity biological waste water treatment in biofilm.

An overview of the literature [2] shows just several studies to have been devoted to low velocity bubble column operation, the hydrodynamic performance analysis of reactors with structured packing being revealed by just a few recent papers. Birrer and Böhm (2004) [1] have reported an overview analysis on the matter. Before them, structured packing parameters have been characterized by Urseanu et al. [3]. Recently, packing hydrodynamics has been studied also by Moustiri et al. [4], Bhatia et al. [5], Nikakhtari and Hill [6], Maldonado et al. [7], and Monsalvo and Böhm [8], The studies are restricted to specific designs. Most of the work concerned non-reactive model systems. Apart from column design, an important group of studies have examined the effect of liquid physical properties [9-12]. From the point of view of diversity of bioreactor objectives, the present state of knowledge points at the importance of studying the performance effects of the various classes of systems separately. These systems include both reactor geometry and fluid physical properties, e.g. pure liquids and their solutions of additives, i.e. low-viscous and highly viscous liquids and liquid phases containing surface-active matter.

In this paper, gas-liquid dispersion in bubble bed packed with fibrous structured mesh material prospective for biofilm application is studied. Gas hold-up and bubble size distribution (BSD) at both growth and non-growth conditions of batches with surface-active and viscous components have been measured and analysed.

2. MATERIALS and METHODS

2.1. Bioreactor setup and packing

All experiments were carried out in a bioreactor column containing the packing. The column and packing pattern are shown schematically in Fig. 1. The packing was made of polymer fiber material 1.2-1.5 mm (*Norten Ltd., France*) produced as polyethylene (PE) and vinyl-acetate (VA) co-polymer, termed PEVA. The geometry comprised a pattern of thermally soldered fibres winded and folded to form a pack of concentric layers filling the circular column cross section. Cases of no-packing, one and two stage packing were examined.

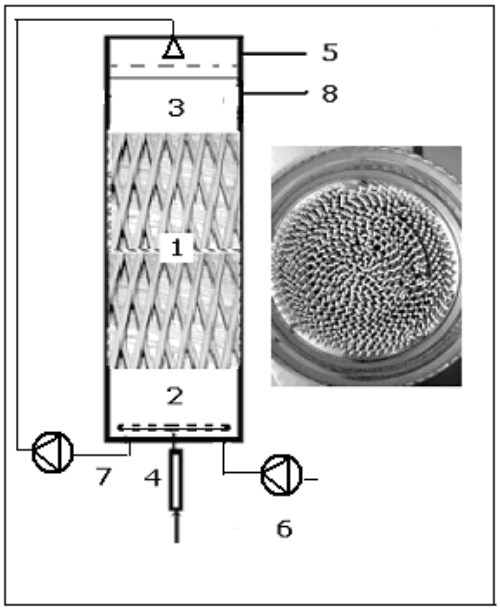


Fig. 1 Schematics of experimental vessel and PEVA packing

A porous plate of diameter 0.06 m mounted on the bottom lid carried out the gas distribution. Gas flow controller and two liquid pumps were used to operate the fluids in up-flow mode with counter-current liquid recycle. Air was used as the gas phase. The bioreactor was equipped with a pH electrode, pH titrator, O_2 probes, and thermometer probe. During the fermentation process inlet and outlet dissolved oxygen concentration, temperature of the medium inside the reactor, air and liquid flow rate were measured and controlled on-line, while pH was measured and controlled off-line.

The experiments were carried out in both continuous and semi batch arrangement at continuous gas flow and at zero or very low liquid feed flow rate. Superficial gas velocity was varied between 0 and 22 mm/s and superficial liquid velocity was varied in the range 0 to 12.6 mm/s involving a liquid recycle 25% to 280 % of liquid feed.

2.2. Model solutions used

Under growth conditions, a biofilm containing *Pseudomonas putida* was cultivated in aqueous saccharose solutions. Under non-growth conditions, analytical grade methanol and saccharose were used to arrange the physicochemical properties of the model solutions. Pure water with surface tension σ = 71.4 mN/m, viscosity μ=1 mPa.s and density ρ = 998 kg.m^{-3} was used as reference. Table 1 contains the physical properties of the systems employed.

2.3. Determination of gas hold-up

Both at growth and non-growth conditions, the gas displacement technique was used. Recent experiments in structured packing by Monsalvo and Bohm [8] showed good reproducibility of the method when checked against pressure tap technique, especially so in the range of gas hold-up 0 to 30%. Changing the liquid inlet and circulation flows and reaching steady state, the valves were turned down and the liquid level corresponding to the liquid flow regime was registered as H_0. Then, the valves were opened and gas flow was let into the system resulting in additional head, the height reaching value H_D. The volume corresponding to this additional liquid head (H_D - H_0) was measured and the percent volumetric gas fraction for this regime was determined as

$$e_G = \frac{H_D - H_0}{H_D} \qquad (1)$$

Table 1. Physicochemical properties of the solutions used.

Concentration	Surface tension – σ, mN/m	Viscosity-μ, mPa.s	Density – ρ, kg.m^{-3}
0 (Water)	71.40	1	998
20 % saccharose	73.00	1.97	1104
40 % saccharose	74.10	6.22	1176
1 g/l methanol	61.02	1	998
2 g/l methanol	54.11	1	997
5 g/l methanol	42.74	1	996
10 g/l methanol	34.87	1	993
2g/l biomass	57.7	0.98	1020

2.4. Determination of bubble size distribution (BSD) and mean bubble size

Bubble size distribution was determined by photographic imaging technique, as follows: Using a electronic digital camera *Nikon GSP 25,* the dispersion field of the flow outlet off the fibrous bed at 5 mm over the bed top surface was photographed and the images were processed by scanning the images electronically on a PC screen to determine number size distributions and average bubble size.

With regard to the distributions obtained, Sauter mean bubble diameter $\boldsymbol{d_s}$ was calculated, as follows:

$$d_{.S} = \frac{\sum_i .n_i d_{bi}^3}{\sum_i .n_i d_{bi}^2} \qquad (2)$$

where $\mathbf{n_i}$ is the number of bubbles of size $\mathbf{d_{bi}}$.

3. RESULTS and DISCUSSION

3.1. The effect of packing height

Gas hold-up was measured first at zero condition of no packing and the data obtained in this case were compared with previous results in conventional bubble columns [10]. The values of gas hold-up obtained at the low gas velocity considered, namely, e_G~7-8% at U_G~0.02 m/s coincided with the reference results, e.g. 4-10 % at U_G ~ 0.01-0.02 m/s.

Further, the case of one (0.13m) and two (0.26m) packing units in the whole range of gas flow rates was examined. The results for gas hold-up and Sauter mean bubble diameter are shown in Figs 2a and 2b, respectively. Gas hold-up is rising almost linearly with packing height (Fig. 2a). The Sauter mean bubble diameter shows a trend towards decrease (Fig. 2b). The effect of the packing is seen as higher dispersion capacity represented by smaller bubbles at rising packing height.

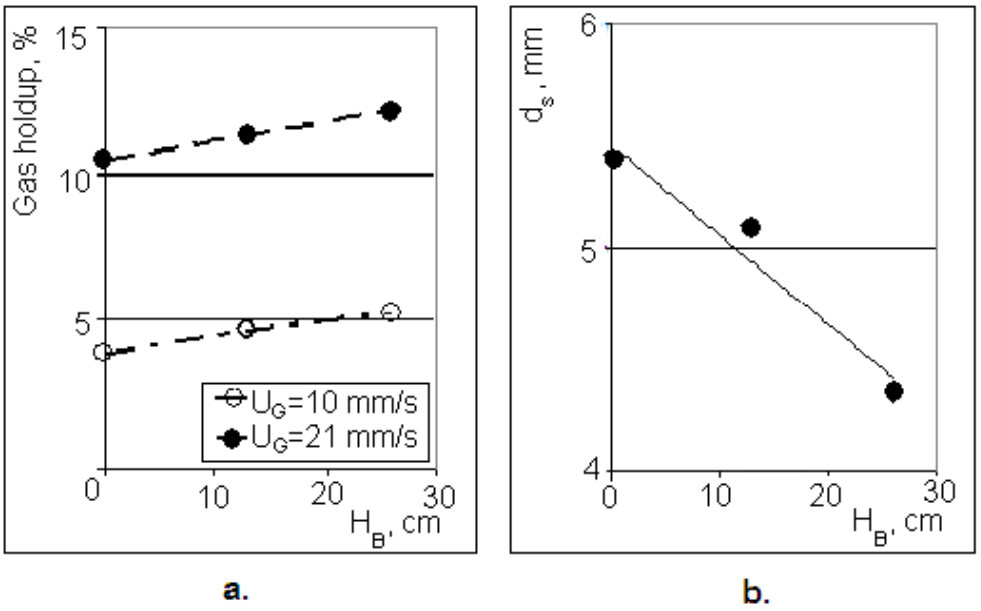


Fig. 2 Gas hold-up and Sauter mean bubble diameter in modular packing 13 and 26 cm

Considering the monotonous gas hold-up effect of packing height, it was inferred that the properties of a single packing unit are representative enough to proceed in the further analysis.

3.2 Gas liquid dispersion at growth conditions

It is extremely important to evaluate the estimates of the major parameters characterising a gas-liquid dispersion at condition of growth. This is because the biological process is strongly dependent on dissolved oxygen and gas hold-up is a major variable component of the mass transfer interfacial area. The other component variable is bubble size. Both parameters were determined in this study. Figs 3 and 4 contain the results.

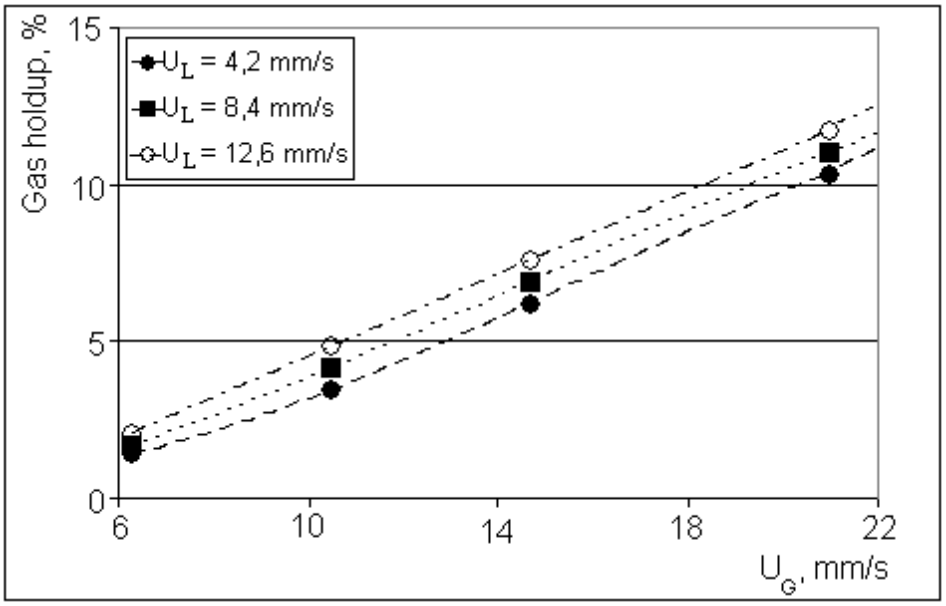


Fig.3 The effect of superficial gas velocity at growth conditions

Gas superficial velocity is the major factor affecting the gas dispersion; its characteristic impact on the reactive gas-liquid flow is illustrated in Fig. 3. A 3-fold increase of U_G was found to cause a 5-fold gas hold-up increase. Liquid superficial velocity being assumed a parameter in this plot, it did not affect gas hold-up considerably: a 3-fold increase in U_L lead to a just 15 % increase of gas hold-up what was within the range of the experimental error. Consequently, the major control on the quality of gas dispersion was related to gas flow rate and the effect of liquid flow on the gas-liquid interface was considered marginal.

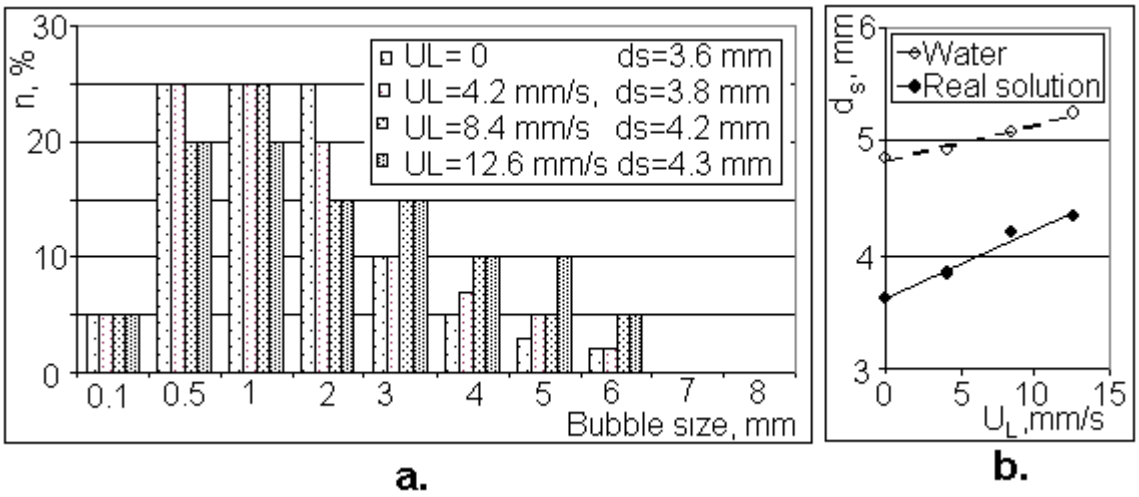


Fig.4 The effect of superficial liquid velocity (U_L) on the Sauter-mean bubble diameter (d_S) at U_G = 6.3 mm/s at conditions of growth: BSD (a), d_s vs. U_L (b)

Fig. 4 shows (4a) the relationship of bubble size distribution (BSD), and (4b) the Sauter mean bubble diameter d_S versus U_L. For the aim of comparison, Fig. 4b shows also the d_S profile in water. As expected, in both cases liquid velocity caused only a minor change on the Sauter mean bubble diameter d_S. For example, the bell-shaped distribution at the end points of the U_L range studied in Fig.4a (being represented by mean bubble size 3.6 and 4.3 mm) showed a difference of less than 1 mm both in real flow and water. However, comparing the plots of growth (ca. 4mm) and non-growth (ca. 5mm) bubble diameters in Fig. 4b, one could infer that the smaller bubbles of the reactive flow represent a larger extent of limited coalescence. Referring to the packing function as a contact unit, one would expect the packing impact to cause break-up and collision. Regarding the highly ionic non-coalescing character of the solution of the bioreactor at conditions of growth, one might expect bubble size to be frozen. In order to clarify this point, additional studies were accomplished using model solutions where the solutes were changed according to pre-selected experimental program. Interest has been invoked in further comparison of gas hold-up at controlled physical properties of the liquid medium. To obtain the effect of the liquid properties, water with controlled amount of additives – alcohols or saccharose – was used.

3.3. Gas-liquid dispersion at non-growth conditions

3.3.1 The effect of U_G for the fibrous packing operating in pure water

Firstly, the effect of superficial gas velocity was examined. Fig. 5 contains the data. Comparable with the one at conditions of growth, a 3-fold increase of U_G yielded a 5-fold gas hold-up rise. The corresponding bubble size distribution in Fig. 6 shows trend towards average bubble size rise (up to 15 %) in the velocity range of U_G~6 to 21 mm/s; With gas velocity rising, it looks likely that the frequency of bubble collisions in the bubble swarm increased leading to increased coalescence. In this sense, the lower the bubble diameter rise, the higher the packing efficiency to disperse the gas in packing presence.

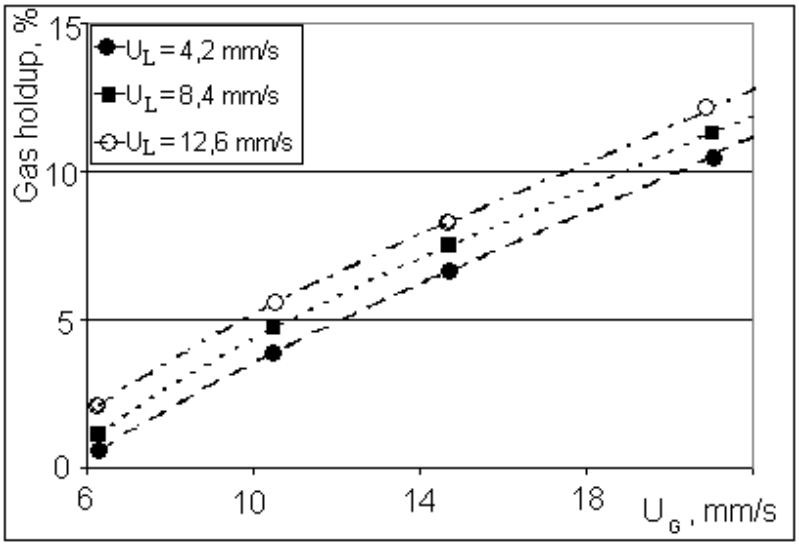


Fig 5. The effect of superficial gas velocity U_G on gas hold-up in water

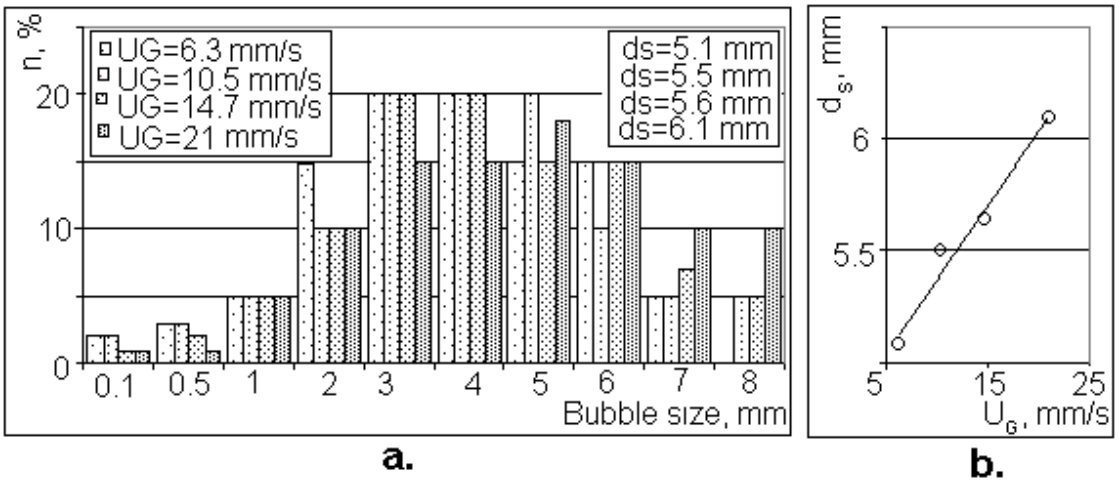


Fig. 6 The effect of superficial gas velocity U_G on bubble size in water: BSD (a), d_s vs. U_G (b)

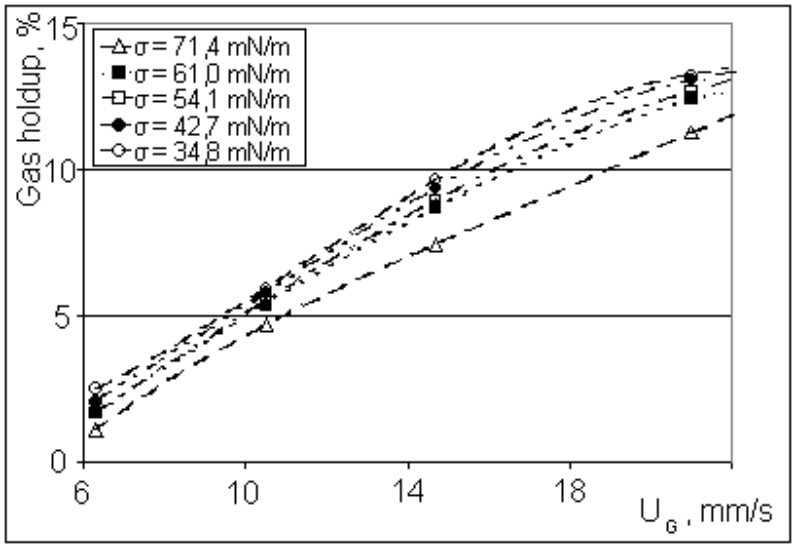


Fig. 7 The effect of surface tension upon e_G

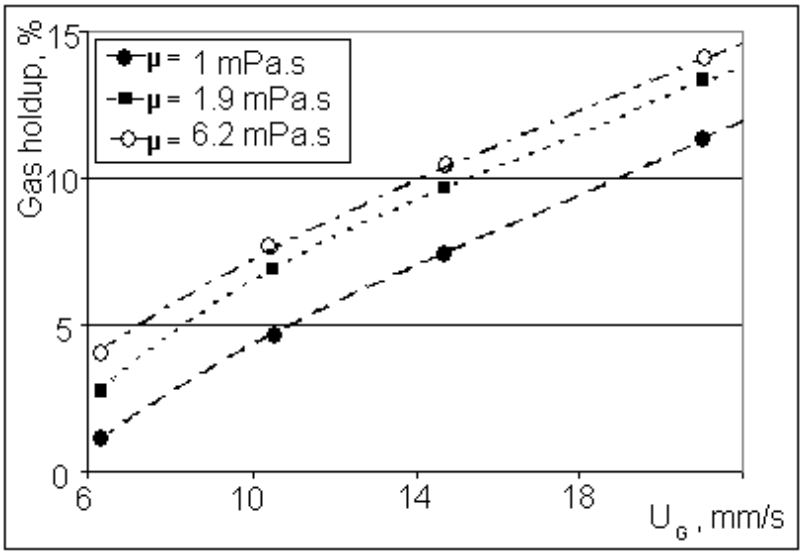


Fig. 8 The effect of viscosity upon e_G

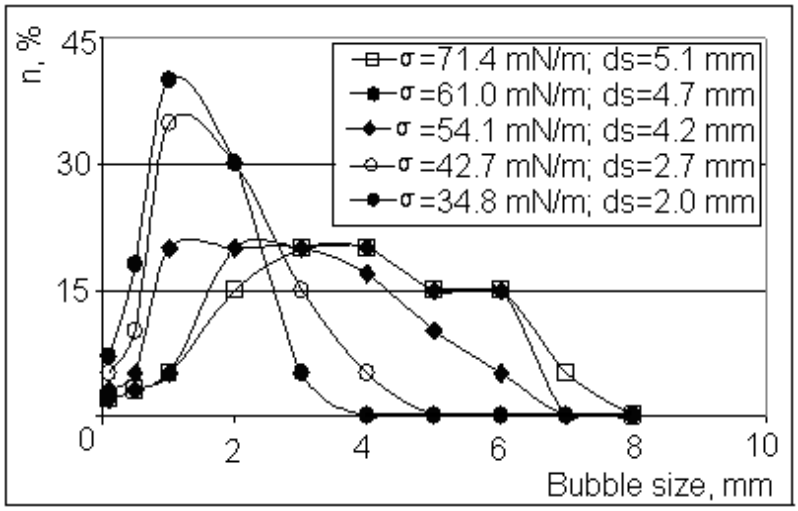


Fig. 9 The effect of surface tension upon BSD

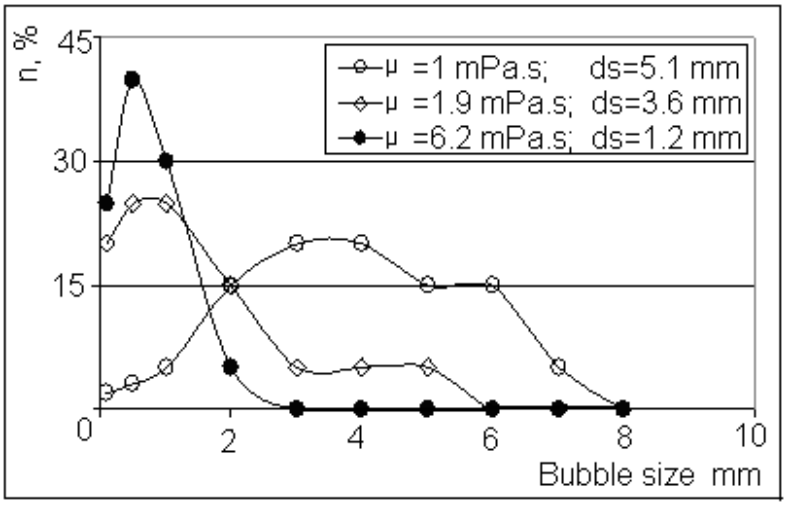


Fig. 10 The effect of viscosity upon BSD

3.3.2The effect of liquid physical properties

The effect of surface tension is shown in Figs 7 and 9, and the effect of viscosity in Figs 8 and 10. It is seen that the presence of surface active additives causes the gas hold-up to increase up to 30 % and $\boldsymbol{d_s}$ to decrease significantly. Evidently, the dispersion capacity of the unit was extended.

Viscosity affects the parameter similarly by decreasing $\boldsymbol{d_s}$ and *i*ncreasing gas hold-up. Qualitatively, the latter effect is similar to the one reported by Zahradnik, Ruzicka and coworkers [10, 12] for conventional bubble columns (without packing). This is however the opposite of what was expected from most of the reference studies (see Kantarci et al., 2009 in [2]). Probably the fact is due to the low values of gas flow velocity examined; one could hardly expect higher flow rates in processing waste water. To our opinion, the result corresponds to the initial rising range of the relationship "gas hold-up vs. viscosity", the gas hold-up having been shown to follow a curve with a maximum [10, 12, 13]. In any case, the reliability of the behaviour e_G *vs* μ observed is confirmed by the size analysis in Fig. 10, as it shows a definite diminishing trend of the Sauter mean bubble size.

3.4. The packing performance in real flow

A summary of reactor dispersion potential, depicted as d_s plots, is shown in Fig. 11. Comparing bubble diameter at growth and non-growth conditions, namely, the reactive (real) nutrient ionic medium (line 1) on one hand, and water with additives – saccharose and methanol, lines 2 and 3, respectively – on the other hand, the packing dispersion performance could be inferred. In Fig. 11, the relationship d_S vs. viscosity (line 2) and d_S vs surface tension (line 3) are plotted together and their intersection with d_S = const (the dotted horizontal line) are shown as points A and B, respectively; both model and reactive flows had equal velocity U_L. By intersection A, one could infer that the real media exhibit as high a dispersion potential (e.g. estimated as d_S) as water with added up to 20 % saccharose. The intersection at B shows that water with added 2 g/dm^3 methanol produced bubble size as large as the real medium did. Because methanol and saccharose are known to suppress coalescence [18-20], the data in Fig. 11 prove that the real flow creates non-coalescing conditions.

Additional information on the packing operation is shown in Fig. 12. It represents an image of bubbles (type 1) captured at the packing channel outlets, and the bubbles (type 2) rising up freely in the bulk. By the difference of bubble size in both cases, one can see clearly that the packing exhibits both a

dispersion function in a way that it ensures small bubble size, and a stabilizing function - in a way that the bubbles remain uniform compared to those performing out of the packing. Consequently, packing performance is effective.

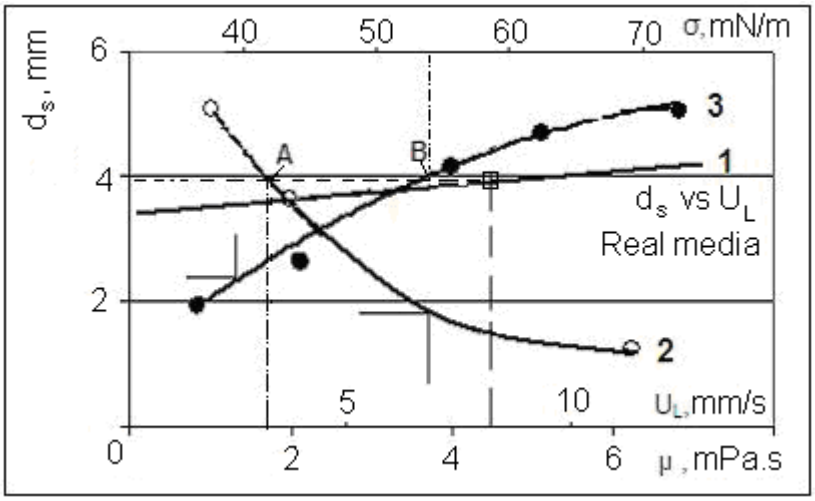


Fig.11 Comparison of bubble size in model and real reactive flow

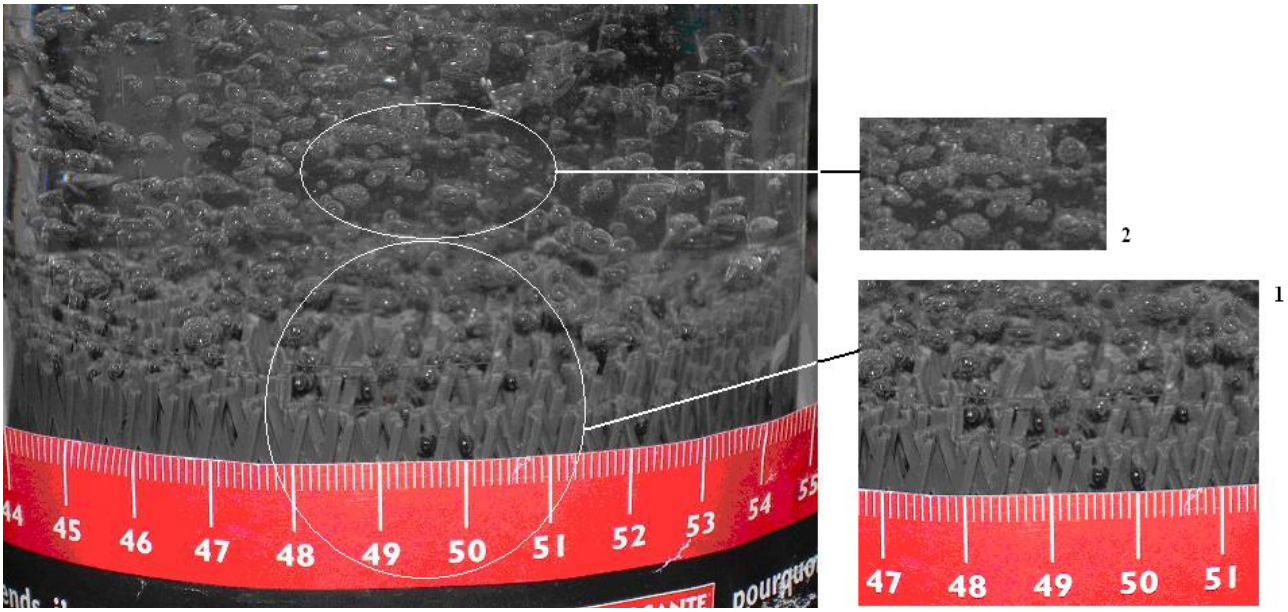


Fig.12 Photograph of gas-liquid dispersion at growth conditions in real flow: views 1 and 2.

3.5. Comparison with reference data of similar systems

In order to verify the dispersion property of the packing, the results of the study were compared with reference data reported by Moustiri *et al.* [4], Bhatia *et al.* [5] and Maldonado *et al.* [7] in **Fig. 13**.

The data sets of the various studies are comparable in a way that they all study gas displacement in packed beds of diameters 0.1-0.15 m at similar superficial liquid velocity, e.g. 12 mm/s, by similar methods and differ within a range of values e_G ~1-2 %. Such differences can be accepted in view of the differences of the packing designs. However, if one considers the data as linear relationships, one could register the deviation of the slope of the curves reported by Moustiri et al. [5] and Maldonado et al. [7]. To our opinion, the reason for such inconsistency could be attributed to large volumetric differences of reported packing porosity. In other words, the major reason for this deviation might be due to the larger bed porosity of the packing studied in [5] and [7] than the one in the case of this study.

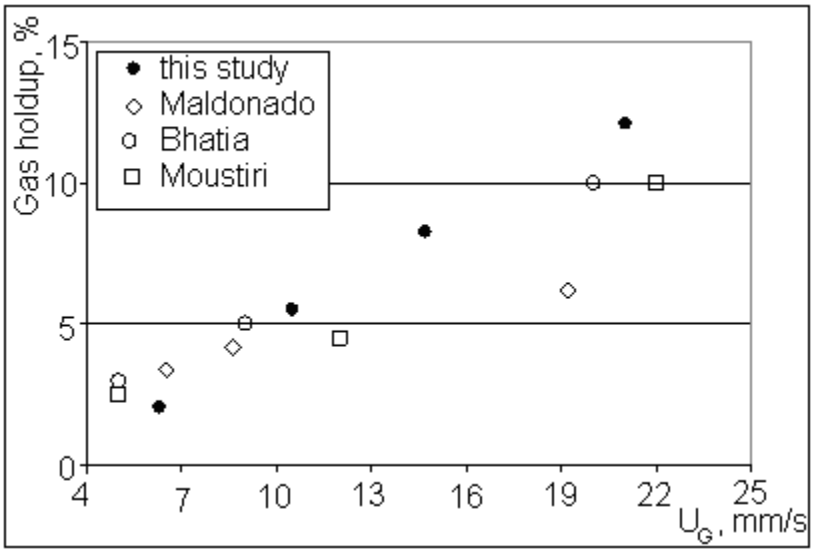


Fig.13 Comparison with reference data

Among the reference data, only Bhatia [5] and Maldonado et al. [7] have reported bubble size data. The Sauter bubble diameter reported by Bhatia et al [5] is 4.5 mm and the one reported by Maldonado et al in [7] is 3-4.6. Both studies reported 1-2 mm lower bubble size compared to the one measured in this study. We have accepted both the values and the techniques employed by these authors as precise enough to believe that the differences in both cases lie entirely within the range of experimental capacity of the techniques employed. Consequently, referring to variation of slopes, we do not believe that the discrepancy in Fig. 12 is caused by variation in bubble size.

3.6. Correlation to operation variables

In order to use the results of this study for extrapolation, an empirical equation was determined, as follows:

$$\varepsilon_G = 1.07 U_G^{0.61} U_L^{0.21} \left(\frac{\mu}{\mu_w}\right)^{0.1} \left(\frac{\sigma}{\sigma_w}\right)^{-0.16} \left(\frac{H_B}{H_{Bo}}\right)^{0.3} \quad (5)$$

Data processing over 70 data sets including 22 sets at growth conditions showed an approximation representative enough by its deviation of 15 %, as evaluated in the parity plot in Fig. 15.

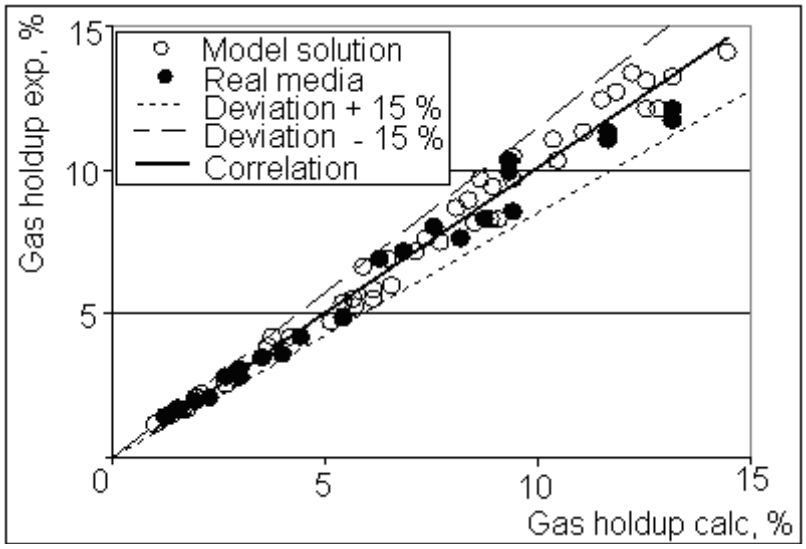


Fig. 14. Comparison between experimental and predicted gas hold-up values.

4. CONCLUSION

The gas-liquid dispersion characteristics of bubble columns packed with fibrous structured material *PEVA* for biofilm application have been studied. Gas hold-up of both reactive and non-reactive batches with surface-active and viscous components was measured. Bubble size distribution was determined. Gas hold-up was found to increase significantly with gas superficial velocity and viscosity and to decrease with surface tension. Liquid superficial velocity showed marginal effect. Gas-liquid flow regime in the packing, as inferred from the specific bubble-to-pore size ratio was classified as bubbly flow. An empirical correlation is proposed that relates gas hold-up with all studied variables. Gas dispersion properties proved to stabilize by the fibrous packing and showed improvement of dispersion parameter by the packing. Assuming geometric similarity of the prototype and the real vessels, the equation at the relevant range of fluid velocities can be used for bioreactor design and scale-up. The results concerning the gas hold-up are shown to be comparable with previous studies of mesh wire packing.

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