Gas hold-up and bubble diameters in a gassed pulsation reactor
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Gas hold-up and bubble diameters in a gassed pulsation reactor

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Abstract

Results of the investigation of gas hold-up in a bubble column (0.13×0.13 m in diameter and 1.6 m high, without baffles) to which pulsations were applied by means of a steel plate located at the tank bottom are presented. The tests were made for water-air and triethylene glycol-air and 0.07% aqueous solution of PAA-air systems at changing frequencies of the plate pulsations in the range from 0 Hz to 33 Hz and at the amplitude from 0.5 mm to 2 mm.

Keywords: pulsating bubble reactor; gas holdup; Bjerknes force.

1. Introduction

Due to pulsations introduced to a column, an increase of mass transfer coefficient in bubble columns was reported already in the 60s of the 20th century. The observed effect caused that a new reactor type, called a pulsating reactor, was constructed. The main aspect of using pulsating (oscillating) reactors is the ability of producing – due to an easy change of pulsations frequency or amplitude – a certain number of bubbles or drops of dimensions optimal from the point of view of mass transfer and residence time in the reactor, using a source of low energy consumption.

The constructions of pulsating (oscillating) reactors can be divided into two main classes: oscillating bubble column reactors (OBCRs) and oscillating baffle reactors (OBRs).

A subject of consideration in this study are oscillating bubble column reactors (OBCRs). From the point of view of design, these reactors are classical bubble reactors equipped with systems producing vibrations which are mounted at the column bottom. Elastic systems (elastic membranes) and rigid systems (e.g. steel plates which behave like a single-piece piston) are used. The main advantage of OBCRs, as compared to baffle reactors, is their simple structure and the fact that along the reactor height the flow of bubbles close to the plug flow is set. Additional backmixing is not initiated. Axial plug flow of bubbles remarkably reduces radial circulation of a mixture and hence in the reactor there is practically no radial phase distribution but only axial one which is very important in some unit processes.

Basing on the literature survey and our own preliminary investigation (Budzynski 2006; Budzynski et al. 2006), it was found that pulsations introduced to the bubble column could cause a significant increase of gas hold-up for two reasons; first because of disturbed gas outflow from a diffuser (a bigger number of small bubbles were formed), and secondly because of the so-called first Bjerknes force which acted opposite to the buoyant force and as result held them up or delayed their outflow.

1.1. The effect of pulsation on gas outflow from an injector to liquid

The size and shape of bubbles in a bubble column depend on the type of gas outflow from an injector (Dziubinski 2005). There are two types of flow: free and jet flow. The criterion, flow rate, at which jet flow appears, was formulated by (Wallis 1966) in the form of relation (1). If this relation is satisfied, then according to the author, the flow should be treated as a jet flow, and if not, there is a free (classical) flow.

$$\frac{V_g\sqrt{r_g}}{\left[gd\left(r_l-r_g\right)\right]^{1/4}} > 1.25\left[\frac{d}{g\left(r_l-r_g\right)R_0^2}\right]^{1/2} \tag{1}$$

This criterion refers to the case when gas is introduced to liquids with physicochemical properties similar to water, it does not take into account rheological properties of a liquid. As proven by Terasaka and Tsuge, Swope and Li, these properties have a significant effect on pressure distribution at the outlet of the injector and hence on the size and shape of formed bubbles.

1.1.1. The effect of pulsations on free gas outflow from the injector

At free flow of the gas from the injector, in the bubble column reactor without pulsations, there is a classical mechanism of bubble growth at the injector outlet. This mechanism is described by many models. (Dziubiński 2005) recommends Kumor-Kuloor or Pińczewski models for gas flow through Newtonian liquids and Li and Swope model for the flow through non-Newtonian liquids. The volume of bubbles formed in these conditions depends mainly on the injector outlet diameter, viscosity and interfacial tension of phases. Due to stable, periodic changes of pressure at the injector outlet, the volume of subsequent bubbles is practically identical. Pulsations introduced into such a system disturb the frequency and amplitude of pressure pulsation at the injector outlet, especially when resonance frequency of pulsations occurs in the system, which can cause break-up of the bubbles and a decrease of their number (Ellenberger et al. 2005; Ellenberger and Krishna 2003; Ellenberger et al. 2003; Ellenberger and Krishna 2002). For instance, as (Knopf et al. 2006b) reported, 10 bubbles per second were generated in the injector of diameter 1 mm at gas flow rate 0.18 cm^3/s, and when this system was subjected to pulsations at the frequency 17.5 Hz, this was practically the resonance value of the system and the number of bubbles increased up to 500 per second.

1.1.2. The effect of pulsations on gas stream flow from the injector

At high flow rates, in the reactor with no pulsations, the classical mechanism of bubble growth at the injector outlet is observed. The volume of bubbles generated in these conditions and hence their diameter distribution is very wide (Knopf et al. 2006b). (Then, several big bubbles are formed; they are of a spherical cap type, or a cloud of bubbles of diameters smaller by one order of magnitude at least.) Introduction of pulsations into this system can change significantly the distribution of bubble diameters as in the case of free flow. In this case, the break-up of bubbles does not depend on their size; all bubbles break up, irrespective of their sizes. This is related to the observed phenomenon of aspirating small bubbles into the injector and next ejecting them along with a portion of liquid to the mixture. Break-up of bigger bubbles can take place as a result of collisions in the portions of liquid which are not ejected (Knopf et al. 2006a).

1.2. The effect of pulsations on the hydrodynamics of bubble flow along the reactor

An increase of gas hold-up in the column and also of mass transfer area can be achieved by dividing the bubbles and holding up or delaying their flow in the column.

Initially, in the 60s, it was presumed that in order to increase the gas hold up, due to pulsations, bubble break-up should be induced thus forcing oscillations which corresponded to natural frequency of bubble oscillations. An equation used to calculate these frequencies for finite column dimensions was proposed by (Bird 1963a; Bird 1963b) on the basis of the known Minnaert's equation of natural bubble frequency in the following form:

$$w_n = \frac{1}{r_o}\left(\frac{3gP_0}{r_l}\right)^{-1/2}\left[1+\left(\frac{r_0}{R}\right)\left(\frac{4L}{R}-1\right)\right]^{-1/2} \tag{2}$$

According to equation (2), the break-up of bubbles of diameter r_0 = 3mm in water caused by vibrations would occur at the frequency about 2000 Hz which is practically unfeasible (Knopf et al. 2006a). So, it can be assumed that the break-up can occur as in the case of flow without pulsations, only due to an increase of bubble volume during flow to the liquid surface and division caused by exceeding of the critical volume (Budzynski 2006; Budzynski et al. 2006) or due to collisions with other bubbles or the column wall.

Although there is no direct experimental evidence of bubble break-up beyond the injector zone under the influence of pulsations many researchers observed over 50% increase of gas hold-up ε at low pulsation frequencies from 5 to 100 Hz. Additionally, it was found that the maximum gas hold-ups occurred at specific frequencies (Bird 1963a; Bird 1963b). For the flow of a small stiff bubble through a non-viscous liquid, (Jameson and Davidson 1966) proposed a simple model which enabled determination of pulsation frequency at which bubble hold-up occurred. In this model the forces acting on small spherical bubbles in a non-viscous liquid, i.e. buoyancy forces, were compared to the resisting force of a medium and Bjerknes force (which acted opposite to the buoyancy force) and the following relation was proposed:

$$\frac{w^4 A^2 r_l h}{2gP_0} = 1 \tag{3}$$

(Jameson 1966) carried out a similar theoretical analysis for the flow of a small bubble through viscous liquid, arriving at the conclusion that the criterion proposed by equation (3) held for Reynolds number $Re_p < 2$, because in this range the influence of viscosity and interfacial tension forces was negligibly small.

(Ellenberger and Krishna 2003; Ellenberger et al. 2003; Ellenberger and Krishna 2002) showed in their studies that the application of low-frequency vibrations in the range 20-100Hz could improve significantly gas hold-up and volumetric mass transfer coefficient. Additionally, they found that for determined operating conditions (bubble column height H, gas flow rate Q_p and gas hold-up ε is not a monotone function of pulsation frequency which shows maximum at harmonic frequencies. In their next study, (Ellenberger and Krishna 2005) basing on theoretical considerations, presented profiles of oscillating changes in pressure and gas hold-up along the column height for five harmonic standing waves of the wavelength λ = 4H, 4H/3, 4H/5, 4H/7 and 4H/9. Based on these considerations they draw a conclusion that along the column height (in the field of the standing wave) the bubbles tend to agglomerate in pressure anti-nodes (Leighton 1990). On the basis of a simple theoretical analysis of averaged changes in gas hold-up, the authors showed and experimentally verified the fact that with a decreasing length of harmonic standing wave occurring along the column height (growing harmonic frequencies) the total gas hold-up increased.

The aim of the study was to prove that
1. continuous phase viscosity had a significant effect on gas hold-up in the pulsating bubble column,
2. an increase of the total gas hold-up in the pulsating bubble column, without baffles, was a synergic effect of pulsations on
a) hydrodynamics of gas outflow from the injector in the immediate zone of its action which might cause break-up of bubbles that during further flow through the column practically did not undergo break up or coalescence induced by pulsations,
b) hold-up or delay of the flow of bubbles due to the Bjerknes force which acts on them (this force is opposite to buoyancy force) mainly in nodes of the harmonic standing wave formed along the column height.

2. Experimental set-up and media

Figure 1 shows a schematic diagram of the experimental set-up. Main part of the measuring system was a glass tank (I-1) on a square base 0.13×0.13 m and 1.6 m high, mounted vertically on a horizontal plate installed inside a stand (I-2). Gas (air) was supplied to the measuring tank by rotameter (II-1) through an elastic pipe with a clamp, to a single nozzle (II-2). The nozzle was mounted 0.2m from the tank base and 0.15m from the plane of the pulsating plate (III-2). Diameters of the nozzles were d_0 = 1, 2 and 4mm. Pressure changes along the tank height were measured by a system of differential manometers connected by elastic pipes to orifices made in the vertical tank wall (I-1) every 200 mm. The orifices were also used to determine gas hold-up ε_{ov} in the column by the volumetric method (i.e. measuring the amount of overflow liquid). The measurements were made for three active heights of the column, counting from the nozzle outlet: H_c= 1.2m, 1.0m and 0.8m.
Pulsations of liquid in the measuring tank (I-1) were induced by a steel plate (III-2) 110 mm in diameter, which was connected to vibrator (III-1) by a lever. Plate (III-2) was connected to the measuring tank by elastic membrane (III-3). The frequency and amplitude of the plate pulsations were set up by controller (III-4). Experiments were carried out at frequency f ranging from 0 Hz to 35 Hz and amplitudes A from 0.5 mm to 2 mm.
The measurements of gas hold-up in the liquid were carried out in the following ranges: changes in the air flow rate Q_p from ca. 50 to 200 cm^3/s. A disperse phase was air from the network and a continuous phase was tap water and triethylene glycol

(viscosity 0.042 Pas) and 0.07% aqueous PAA solution (n = 0.7[-], k = 0.023 Pas^n). The measurements were made at ambient temperature, 21-24°C. During the measurements photographs of particular column sections were taken which enabled estimation of the amount, size and bubble diameter distribution along the column.

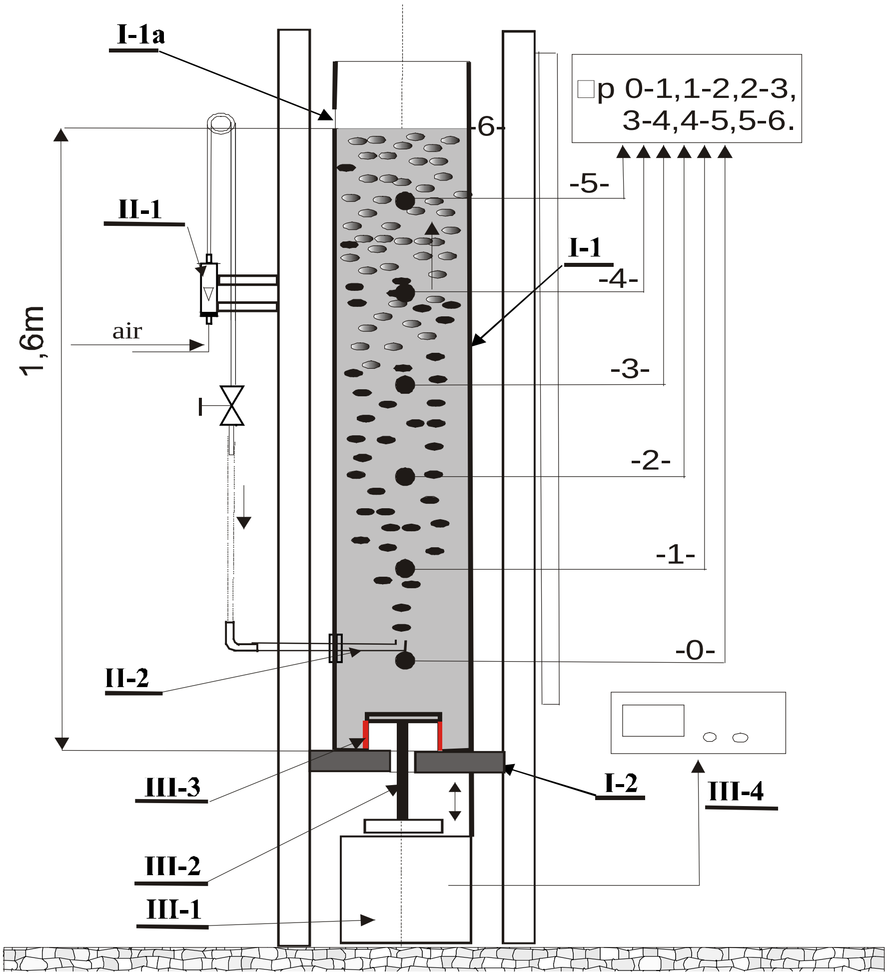


Fig. 1.Schematic diagram of the experimental set-up

3. Results and discussion

Basing on the experimental results, the following conclusions were drawn:

a)The mean gas hold-up in the pulsating bubble column at resonance frequency (harmonic standing wave) was significantly higher than in the column operating at non-resonance frequencies. In the tested range of changes in the disperse mixture height H and the frequency and amplitude of pulsations A, two resonance frequencies were found. For instance, for the water-air system, the resonance frequency at pulsation frequency f^* 11 Hz and 33 Hz was reported, cf. Fig. 2.

b)When the column works at resonance frequencies, gas bubbles (local increase of gas hold-up) concentrate along the column height in places where the arrow of a standing wave occurs (in pressure anti-nodes). No break-up or coalescence of the bubbles was observed. Figures 4a-b and 5a-c show some photographs taken along the column height where the first (I) and second (II) resonance frequency was observed for water-air system Q_p = 165 cm^3/s, H_c = 1 m, R_0 = 1 mm, A=1 mm. The effect of bubble concentration in the arrows was enhanced with an increase of gas flow rate per unit volume and with the growth of plate pulsation amplitude, cf. Fig. 3.

c) Basing on the analysis of photographic documentation in the tested range at resonance frequencies, a significant increase of the number of formed bubbles with a simultaneous decrease of their diameters in reference to the flow without pulsations, an increase from 100 to 300%, at the same flow rate was observed, cf. Fig. 6a-b.

d) Resonance frequencies were also reported in experiments with the glycol-air system, and 0.07% aqueous PAA solution-air, however, gas hold-up was several times lower than in the air-water system at the same pulsation parameters, cf. Fig. 7.

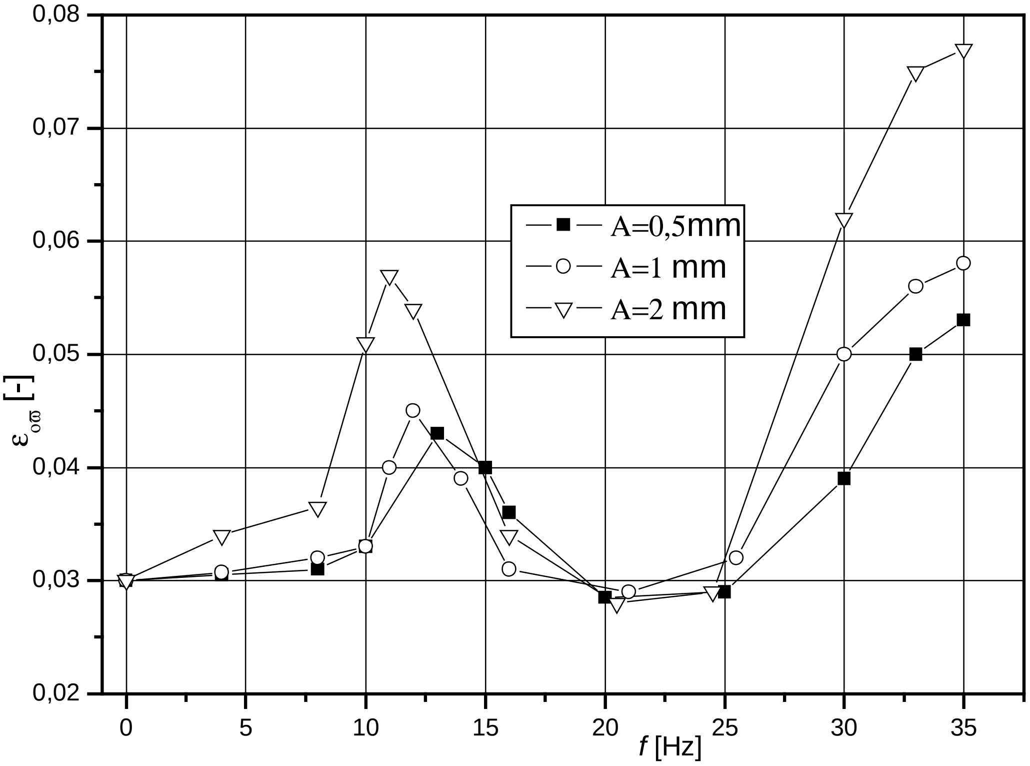


Fig. 2. The effect of pulsation frequency *f* (plate) on gas hold-up ε_{ov} in the bubble column for water-air system, Q_p = 165cm^3/s, H = 1m, R_0 = 1mm.

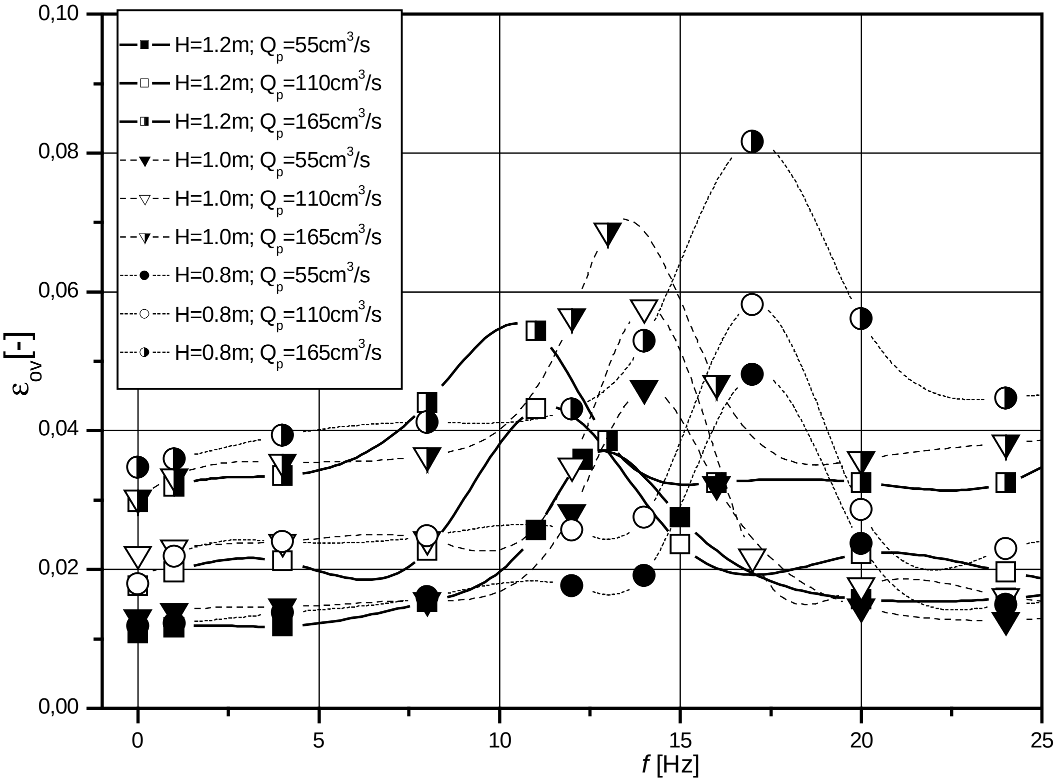


Fig. 3. The effect of pulsation frequency *f* (plate) on gas hold-up ε_{ov} in the bubble column for water-air system, $Q_p = 165cm^3/s$, $H_c = 1m$, $d_0 = 2mm$.

a)

b)

Fig. 4. Photograph (fragment) of a bubble column, a) in “arrow I” at the surface, b) in “node”, at the first resonance frequency, f^* =11Hz

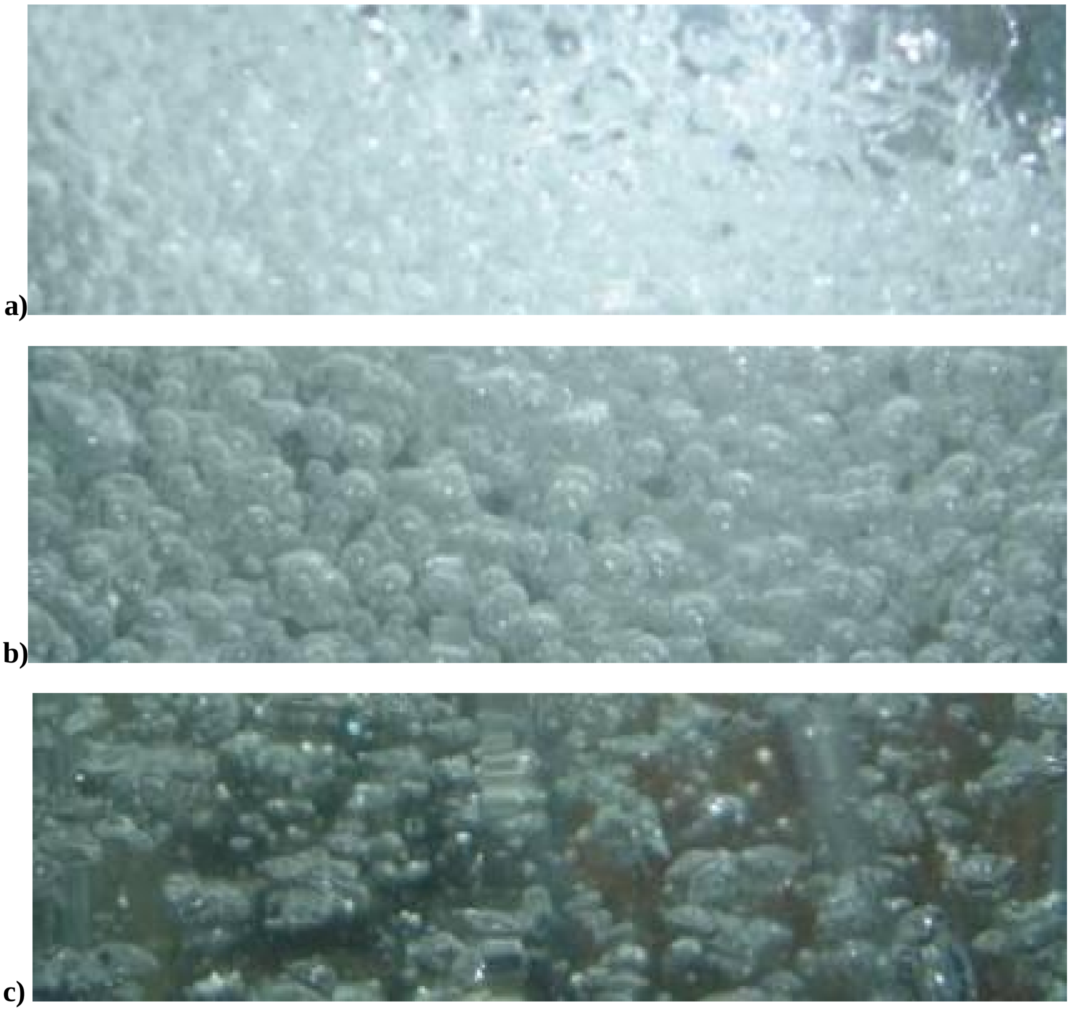
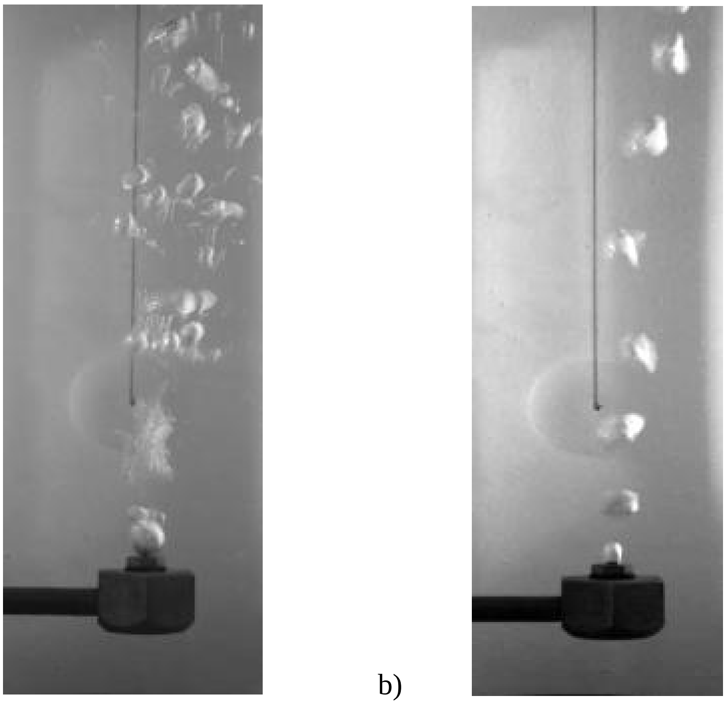


Fig. 5. Photographs (fragments) of a bubble column; a) in “arrow II” at the surface, b) in “arrow I” c) in “node”, at the second resonance frequency, f^* =33Hz



a) b)

Fig. 6. Photograph (fragments) of the bubble column at the injector outlet; a) with pulsations, second resonance frequency, f^*=33Hz, Q_p = 165cm^3/s, H_c = 1m, R_0 = 2mm., A = 2mm, b) no pulsations,

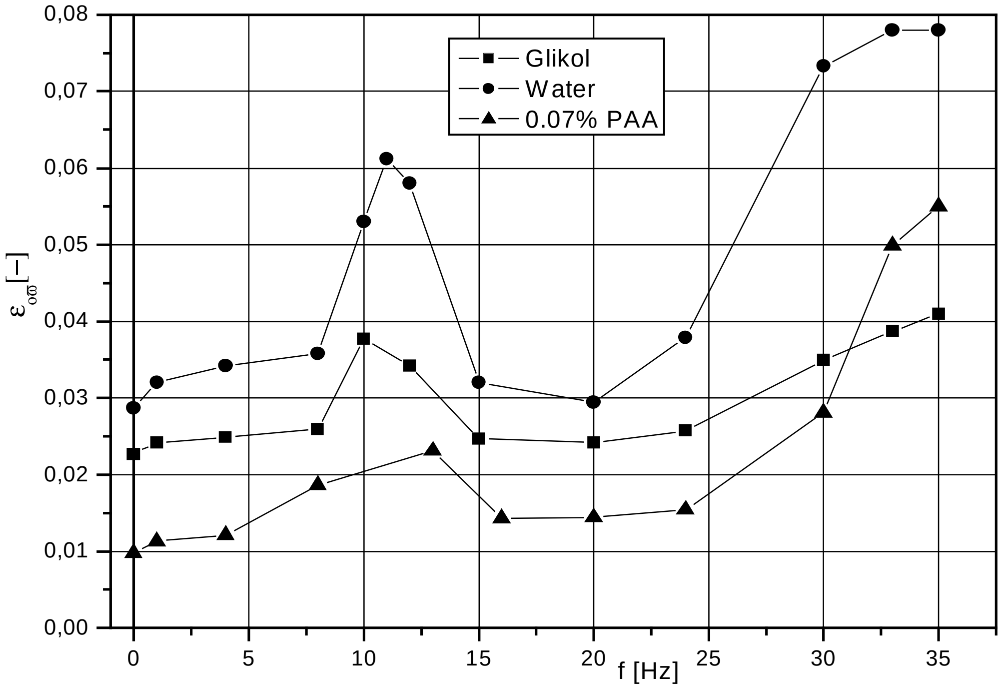


Fig. 7. Comparison of the effect of pulsation frequency f (plate) on gas hold-up ε_{ov} in the bubble column water-air, glycol-air and 0.07% aqueous PAA-air solution, Q_p = 165 cm^3/s, H_c = 1m, R_0 = 2mm., A = 2mm

4. Summary

1. Experimental results confirm the principle presented by (Ellenberger and Krishna 2005; Ellenberger and Krishna 2003; Ellenberger et al. 2003; Ellenberger and Krishna 2002) concerning the harmonic standing wave along the pulsating bubble column. The maximum gas hold-up was observed at_resonance frequencies and agglomeration of bubbles in the arrows of standing waves was reported even in the tested range of low values of frequency changes f from 1 to 33 Hz.

2. When analyzing results available in literature, including our own published results (Budzynski 2006; Budzynski et al. 2006) and the present research, it should be claimed that the growth of gas hold-up in the pulsating bubble column without baffles, is a result of two phenomena:

a) The interaction of pulsations on the hydrodynamics of gas outflow from the injector and in the immediate zone of its activity which can cause break-up of bubbles, which in higher parts of the column do not undergo further break-up or coalescence induced by pulsations.

b) As a result of hold-up or delay of bubble flow in the arrows of harmonic standing wave, at pulsations of resonance frequency, due to the action of Bjerknes force in these points.

3. A significant influence of continuous phase viscosity on gas hold-up was observed. This seems to confirm a very limited range of gas bubble outflow proposed by (Jameson and Davidson 1966) and Jameson (1966). The observed significant decrease of gas hold-up with an increase of continuous phase viscosity at practically the same resonance frequencies, cf. Fig. 7, justifies the application of second-order underdamped process with inertial viscoelastic behavior in the description of response of the liquid-plate-pulsator system (Knopf et al. 2006a).

Nomenclature

λ – wavelength, m
f – frequency, Hz
f^* – resonance frequency, Hz
h – mean depth of bubble, m
A – amplitude, m
H – height of gas-liquid in the column, m
L – mean depth of bubble below surface, m
P_0 – pressure inside bubble, N/m^2
Q_p – gas volumetric flow rate, m^3/s
r_o - bubble radius, m
R – radius of column, m
Re - Reynolds number, $Re=fAR_0/\nu$
R_0 – orifice radius, m
V_g – nozzle gas velocity, m/s
e_{ov} – height-averaged gas hold-up, dimensionless
g – specific heats ratio of gas (=1.4 for air), dimensionless
ν – kinematics viscosity, m^2/s

r_l – liquid density, kg/m^3
r_g – gas density, kg/m^3
d – surface tension, N/m
w – frequency of oscillation, rad/s
w_n – bubble system natural resonance frequency, rad/s

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