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# Gas hold-up analysis in an unsteady stirred vessel by means of infinite series

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The use of a liquid level sensor made it possible to measure changes in gas hold-up over time in a stirred tank during unsteady mixing. These results were subjected to Fourier time series analysis and a model of gas hold-up changes in time was proposed. It allowed one to determine the model value of gas hold-up, which can be useful for characterizing gas hold-up during unsteady mixing and as a comparison to gas hold-up during steady mixing. The characteristic frequency was also determined, which corresponds to about twice the oscillation frequency. Model gas hold-up values for coalescing and non-coalescing systems were compared. Moreover, the change of the gas hold-up at constant maximum stirrer rotation frequency and variable gas flow rate for different oscillation frequencies was investigated.

**Keywords:** unsteady mixing, gas hold-up, non-coalescing system, stirred vessel.

## INTRODUCTION

At a constant gas flow rate, as the impeller speed increases, the state of dispersion in a stirred tank changes from flooding through gas loading and full dispersion to recirculation[1]. Since in unsteady mixing during one cycle the impeller speed changes from 0 up to $N_{max}$, almost all dispersion states occur. The gas hold-up in a stirred vessel is a key parameter frequently described in literature[2–5] since it provides valuable information on the mixing of gas-liquid systems[6]. It is defined as the ratio of the volume of gas retained in the liquid to the volume of the liquid ($\varepsilon$) or as the ratio of the volume of gas retained to the volume of the dispersion ($\phi$). It indicates the amount of gas in the two-phase mixture, which is related to the efficiency of the impeller in dispersing the gas. The ability of the impeller to increase interfacial area is especially important during mass transfer in such systems.

The most common way to measure the gas hold-up is by the volumetric method. It consists of measuring the change in the system's volume under the influence of gas dispersion through the impeller. This parameter could be assessed visually[2, 7], which is a major inconvenience and is affected by a large measurement error due to the dynamics of the system. It is very important, especially in unsteady mixing. In such mixing[5, 8], the gas hold-up changes with time and with impeller speed over time. One technique for determining gas hold-up during unsteady mixing is to visually determine the maximum and minimum values[8, 9]. In addition to this technique, some researchers[5, 10–12] used the manometric method proposed by Robinson and Wilke[13] in which the gas hold-up was calculated based on the drop in the hydrostatic pressure difference, taking into account the dynamic pressure correction. This correction assumed that the dynamic pressure in the gas-liquid dispersion and gas-free liquid is the same. Both the hydrostatic pressure difference and the dynamic pressure are time-varying for unsteady mixing. They are recorded during separate measurements, which makes it difficult to determine the change in gas hold-up over time, and operating with average values is also subject to error. Information on the course of changes in gas hold-up could provide valuable information on phenomena occurring during unsteady mixing, e.g. flooding of the impeller.

Several new techniques have been considered that could measure the degree of gas hold-up, especially during unsteady mixing[14, 15]. The most effective among those considered turns out to be the measurement with the eTape Milone resistance sensor and the software developed for this purpose.

The aim of the work was to develop a methodology for determining gas hold-up during unsteady mixing using a liquid level sensor and registering gas hold-up changes over time as an alternative to other techniques.

## MATERIALS AND METHODS

The experimental set-up (Fig. 1) consisted of a flat-bottomed tank (I) with a diameter of $D$ = 0.29 m. There was a possibility to install in it a gas sparger (II) with a diameter of $d_b$ = 0.085 m. A Rushton turbine with a diameter of $d$ = 0.1 m was used as the impeller (III). Sensor AT's Mt2 torque meter (IV, V) allowed for torque and rotation frequency measurement. The shaft

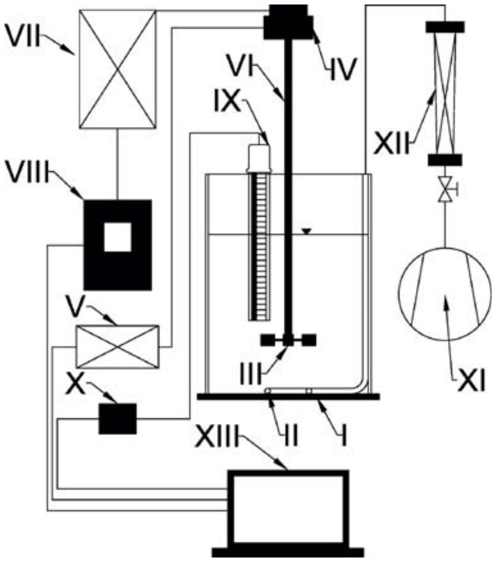

**Figure 1.** Experimental set-up: vessel (I), gas sparger (II), impeller (III), torque sensor (IV) torque meter (V),, shaft (VI), electric motor (VII), inverter (VIII), liquid level sensor (IX), liquid level meter (X), compressor (XI), rotameter (XII), PC (XIII)

(VI) was driven by an Elektrim SF400L4A electric motor (VII) controlled by a Schneider Electric Company pDrive MX Eco inverter (VIII) and MatriX computer software, which allowed for programming changes in the rotation frequency over time. In addition, the eTape Milone liquid level sensor (IX) was mounted in the tank, which allowed for recording the change in the liquid level over time with a high sampling frequency in the range from 10 to 300 Hz.

The liquid level at a given moment was converted to gas hold-up using the formula.

$$\varepsilon = \frac{\Delta H}{H_0} \tag{1}$$

where $\Delta H$ is the change in height of the system and $H_0$ is the height of the liquid phase which was $H_0 = 0.29$ m.

The tests were carried out for the air-water and air – 0.2 M NaCl solution systems in the range of air flow rates ($Q$) from 0.5 to 3 $m^3$/h (superficial gas flow velocity ($w_g$) from 0.0021 m/s to 0.0126 m/s). The forward-reverse mixing was carried with the course of the triangular wave for the impeller speed ($N$) in time ($t$), in the oscillation frequency ($f$) range from 0.115 Hz to 0.46 Hz, for the maximum impeller speed ($N_{max}$) from 5 to 14 rpm.

The obtained results of the gas hold-up were subjected to Fourier time series analysis using the Hamming window in the TIBCO Statistica 13.3 software. The periodogram value was determined using the equation[9]:

$$P_k = \left(\sum_{N=1}^{H} a_N^2 + b_N^2\right)\frac{N_s}{2} \tag{2}$$

where $N_s$ is the length of the Fourier series, and $a_N$ and $b_N$ are the characteristic coefficients of the series.

Model parameters were determined using Mathworks Matlab R2022a software by nonlinear least squares method and Levenberg-Marquardt algorithm.

## RESULTS AND DISCUSSION

In the first stage of the research, the data obtained from the liquid level sensor was subjected to time series analysis. The characteristic harmonic frequencies were identified by analysing the periodogram values. Due to the appearance of these characteristic frequencies, a model of change in gas hold-up over time was proposed in the following form:

$$\varepsilon(t) = \varepsilon_0 + a_1\cos(2\pi\omega t) + b_1\sin(2\pi\omega t) \tag{3}$$

where $\varepsilon_0$ is the model gas hold-up value, $a_1$ and $b_1$ are constants, and $\omega$ is the characteristic frequency of the model.

In Figure 2, Figure 3, and Figure 4, the results of the time series analysis for impeller speed, gas hold-up, and model (3) for 3 different oscillation frequencies are presented. The frequencies 1f and 2f are marked. Local maxima of periodogram values appear at frequencies equal to multiples of the oscillation frequency. The highest periodogram values for gas hold-up and model (3) are observed for a frequency close to twice the impeller frequency ($I_2 \approx 2f$). $I_2$ frequency is considered as the second harmonic frequency of changes od liquid level and as well gas hold-up. During the torque analysis for unsteady mixing, odd frequencies usually appear[9], but in this case, even frequencies were observed.

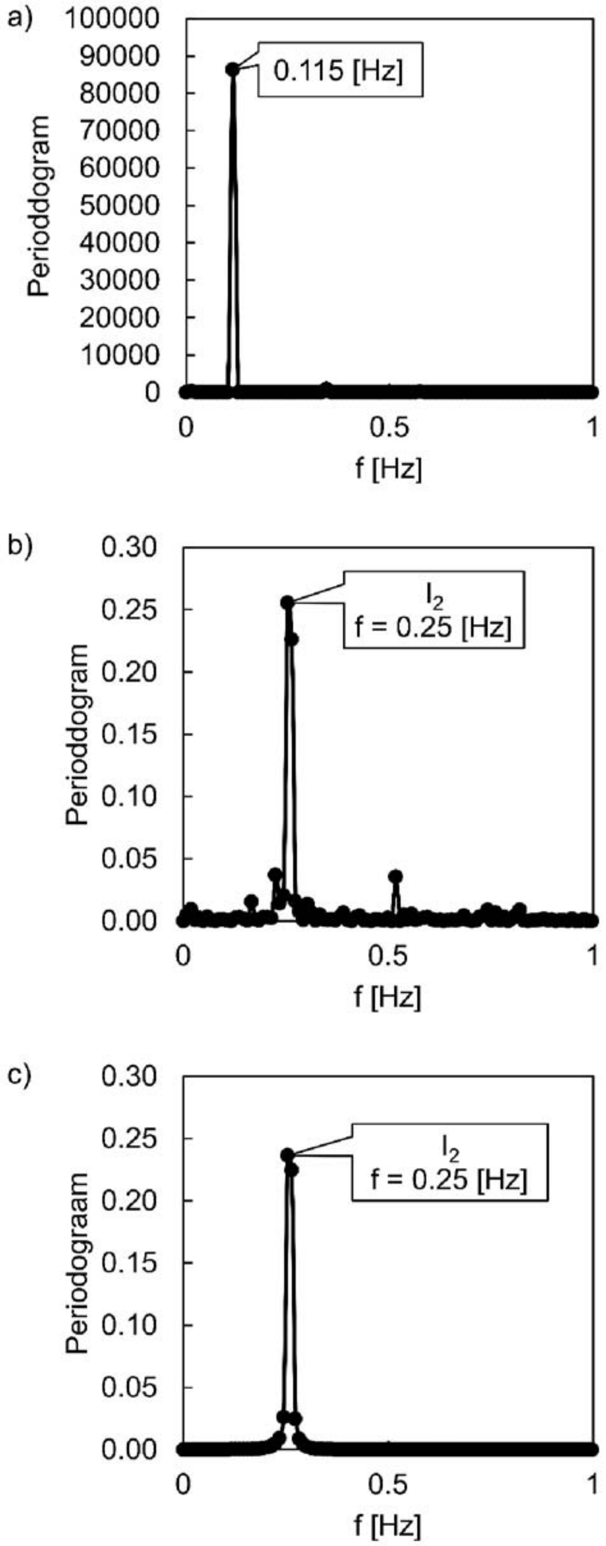

**Figure 2.** Periodogram for: impeller speed (a), gas hold-up (b) and model (c) f=0.115 [Hz]

Figure 5 shows an example graph of gas hold-up versus time during unsteady mixing recorded with the eTape sensor. There are clear changes in the gas hold-up. The use of the sensor makes it possible to register gas hold-up in time even for high oscillation frequencies. The values calculated based on model (3) were also plotted on the graph. Table 1 shows the values of coefficients, errors, p-values, confidence bounds as well as R-square, the sum of squares due to error (SSE), and root mean squared error (RMSE) for this fit. Due to the dynamics

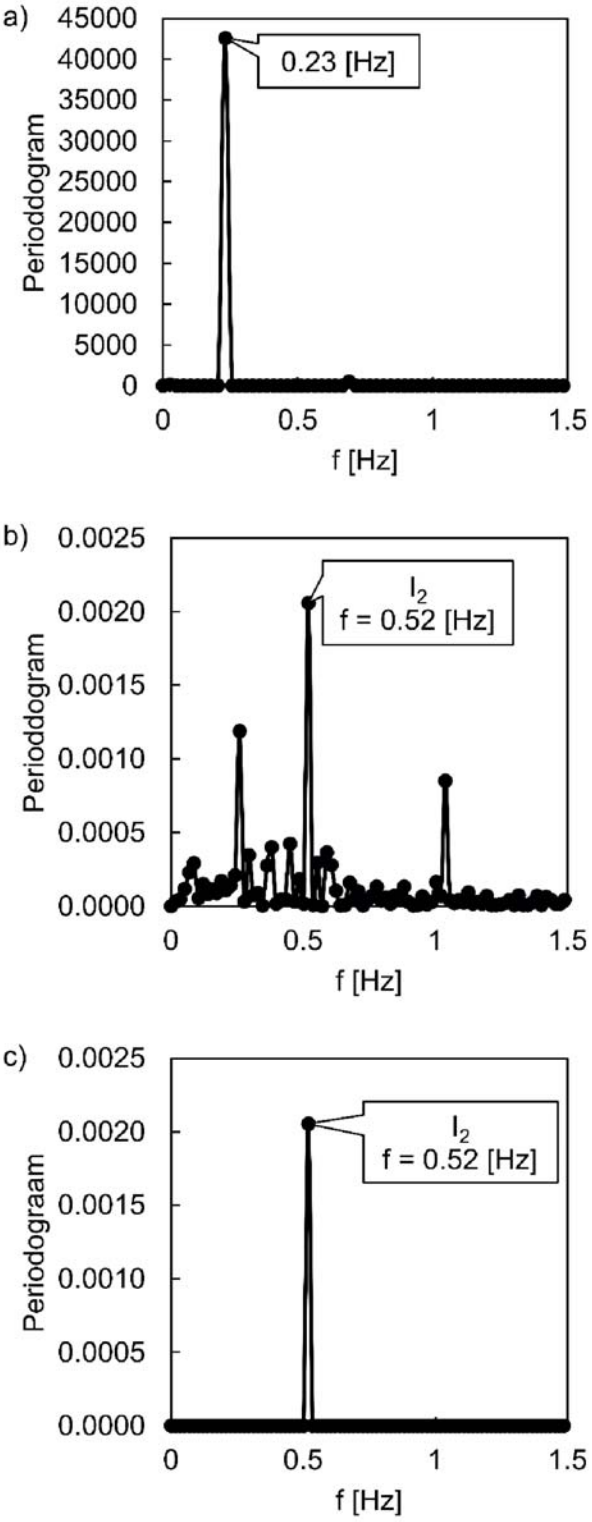

**Figure 3.** Periodogram for: impeller speed (a), gas hold-up (b) and model (c) f=0.23 [Hz]

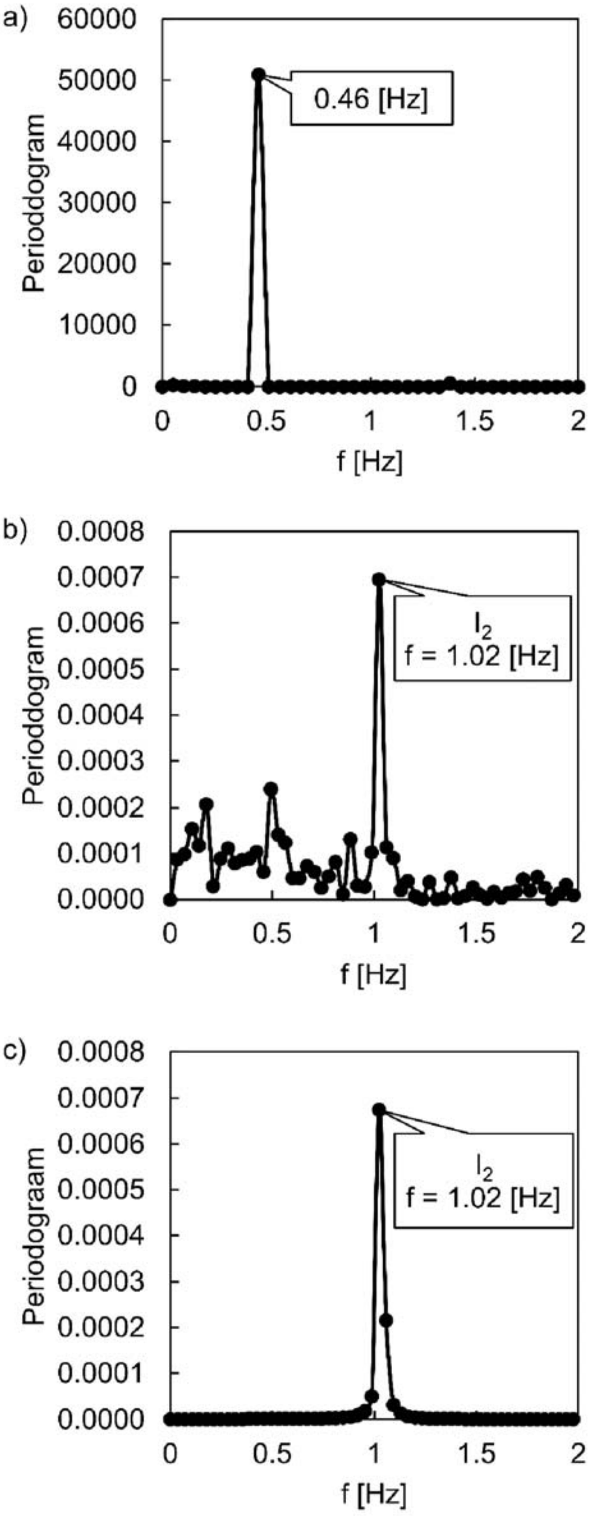

**Figure 4.** Periodogram for: impeller speed (a), gas hold-up (b) and model (c) f=0.46 [Hz]

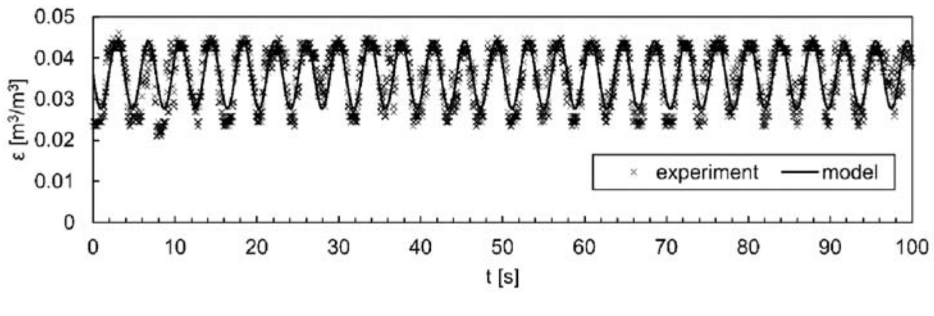

**Figure 5.** Exemplary gas hold-up course during unsteady mixing and its fit to the model (3)

**Table 1.** The statistical results of the fit shown in Figure 5

| | coefficients | error | p-value | confidence bounds | | $R^2$ | SSE | RMSE |
|---|---|---|---|---|---|---|---|---|
| $\varepsilon_0 \left[\frac{m^3}{m^3}\right]$ | 0.0355 | 0.0001 | $<10^{-6}$ | 0.0354 | 0.0357 | 0.7052 | 0.0777 | 0.0038 |
| $2\pi\omega\ [Hz]$ | 1.6208 | 0.0008 | $<10^{-6}$ | 1.6193 | 1.6223 | | | |
| $a_1$ | –0.0014 | 0.0002 | $<10^{-6}$ | –0.0018 | –0.0009 | | | |
| $b_1$ | –0.0084 | 0.0001 | $<10^{-6}$ | –0.0086 | –0.0082 | | | |

of the system and the undulation of the liquid surface during unsteady mixing, we found the obtained parameters to be satisfactory. Figure 6 shows the residual normality plot, which indicates that the residuals are normally distributed.

Based on the previously obtained periodograms, it was established that the characteristic frequency of the model corresponded to approximately twice the oscillation frequency (2f). In addition, the proposed model allows one to obtain the value of $\varepsilon_0$, which can be used as a value characterizing the gas hold-up for unsteady mixing. This will allow the gas hold-up to be compared both during unsteady mixing and to values normally used for steady mixing.

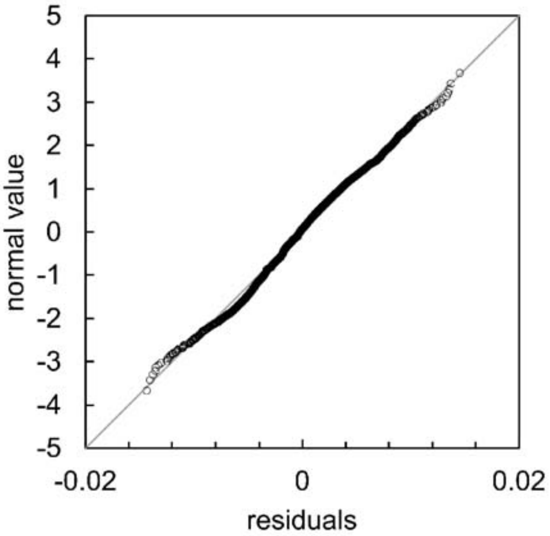

**Figure 6.** Residual normality plot for the fit shown in Figure 5

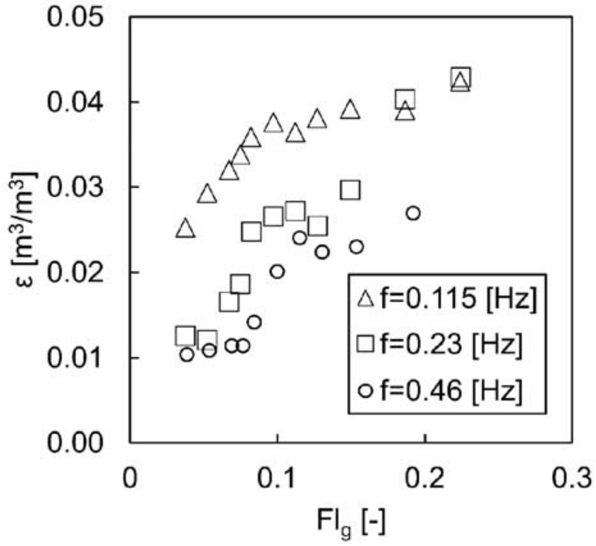

**Figure 7.** Comparison of model gas hold-up $\varepsilon_0$ depending on gassed power input per unit volume Pg/V for coalescing and non-coalescing systems for different gas flow rates Q

Figure 7 shows an exemplary comparison of gas hold-up depending on the gassed power input per unit volume for coalescing (air-water) and non-coalescing systems (air-salt solution) obtained for different gas flow rates Q. As expected, for all measurement series, an increase in gas hold-up is observed with an increase in gassed power input per unit volume as well as an increase in gas flow rate. The effect of the addition of NaCl as a substance preventing the coalescence of gas bubbles is also visible. The addition of salt results in a more rapid increase in gas hold-up as gassed power input per unit volume increases and higher gas hold-up values are obtained.

Figure 8 shows the obtained dependence of $\varepsilon_0$ on the gas flow number $Fl_g$ for the air-water system in the range of oscillation frequency from 0.115 Hz to 0.46 Hz. With a constant maximum impeller speed of about 6 rps, the gas flow rate was changed in the range from 0.5 $m^3$/h to 3 $m^3$/h. The conducted research shows that the value of $\varepsilon_0$ increases with the gas flow number and decreases with the increase of the oscillation frequency. In other works[8, 9] the maximum and minimum gas hold-up values were determined. These values, in turn, increased with increasing oscillation frequency. The difference in the effect of the oscillation frequency on these parameters

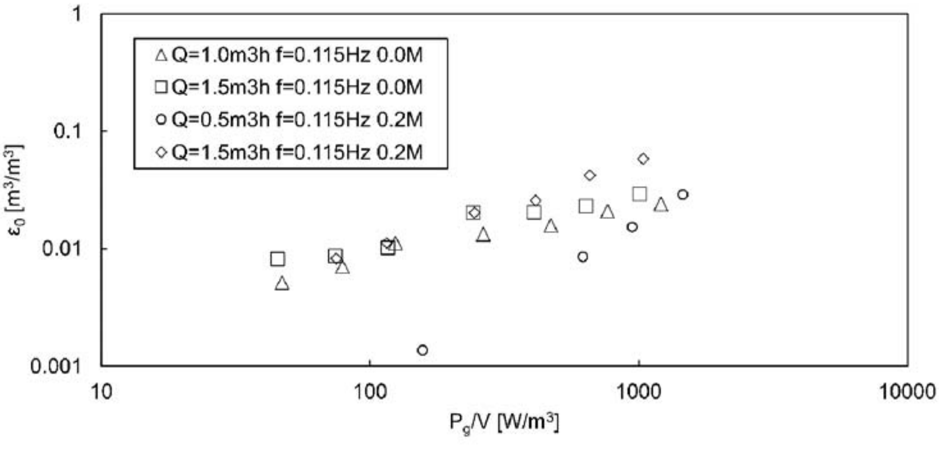

**Figure 8.** Dependence of model gas hold-up $\varepsilon_0$ on gas flow number $Fl_g$ for air-water system

may be due to a different measurement method. The significant undulation of the liquid surface significantly affects the minimum and maximum values determined by the visual method. Therefore, the technique of measuring the change in gas hold-up over time and determining $\varepsilon_0$ using model (3) should give more complete information about the course of gassing, and not only extreme values, which may allow drawing other conclusions.

Four ranges of changes in the value of $\varepsilon_0$ with $Fl_g$ were observed, occurring sequentially from the lowest (I) to the highest (IV) $Fl_g$ values. In Range I there is a slight increase in the value of $\varepsilon_0$ with $Fl_g$. The next, Range II, includes a sharp increase in the value of $\varepsilon_0$ with $Fl_g$ until reaching a local maximum. In Range III there is a slight decrease in the value of $\varepsilon_0$ with increasing $Fl_g$ until reaching a local minimum at about $Fl_g = 0.1$, which corresponds to a gas flow rate of about 1.3 $m^3/h$. In Range IV there is a further increase in the value of $\varepsilon_0$ with the increase of $Fl_g$. The rate of this increase is lower than that of Range II. In the case of the frequency $f = 0.115$ Hz, the occurrence of the Range I was not observed.

During the forward-reverse mixing running in accordance with the triangular wave, it is assumed that impeller flooding always occurs to some extent, regardless of the oscillation frequency and the maximum value of the impeller speed. The appearance of $\varepsilon_0$ drops in Range III may indicate flooding in the entire range of impeller speed changes and a further increase is caused by increasing gas flow rate.

## CONCLUSIONS

Overall, the eTape sensor provided valuable information on gas hold-up during unsteady mixing. However, the analysis of the periodograms did not allow to identify the flooding of the impeller during unsteady mixing and further studies are required. The eTape sensor, due to its design and the method of measuring the change in the height of the liquid level in the tank, causes many difficulties due to the dynamics of the system during mechanical mixing, in particular unsteady mixing. Further development of the technique could be focused on multi-point or entire perimeter measurement of the tank, which would allow for more accurate data.

## NOMENCLATURE

$a_1$ – model parameter
$a_N$ – characteristic coefficient of the Fourier series
$b_1$ – model parameter
$b_N$ – characteristic coefficient of the Fourier series
$D$ – tank diameter, m
$d$ – impeller diameter, m
$d_b$ – sparger diameter, m
$f$ – oscillation frequency, Hz
$Fl_g$ – gas flow number
$H_0$ – height of the liquid, m
$l_2$ – frequency close to twice the impeller speed, Hz
$N$ – impeller speed, rps
$N_s$ – length of the Fourier series
$P_g$ – gassed mixing power, W
$P_k$ – periodogram value
$Q$ – gas flow rate, $m^3/h$
$V$ – liquid volume, $m^3$
$t$ – time, s
$\omega$ – model characteristic frequency, Hz
$\Delta H$ – change in height of the system, m
$\varepsilon$ – gas hold-up (to the volume of the liquid), $m^3/m^3$
$\varepsilon_0$ – model value of gas hold-up, $m^3/m^3$
$\varphi$ – gas hold-up (to the volume of the dispersion), $m^3/m^3$

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