

Parameters characterising the pulsing flow for cocurrent flow of gas and foaming liquid in a pressurised trickle-bed reactor

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## **Parameters characterising the pulsing flow for cocurrent flow of gas and foaming liquid in a pressurised trickle-bed reactor**

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### **Abstract**

The aim of the present work was to experimentally determine the parameters characterizing the pulse flow of liquids through bed i.e. the frequency of pulsation and the velocity of pulses travelling along the bed and next to work out the correlation equations enabling their assessment. The research was carried out in a pressure reactor (pressures up to 2 MPa) having 0.05 m in diameter and being 1.5 m long. Physicochemical properties of the gas and liquid phases were changed in the research and the concentrations of water solutions of aliphatic alcohols were chosen in such a way so that the systems would form foams of various lifetime as a result of their contact with the gas in packing. The experimental data which were obtained as a result of the research were correlated depending on the operational parameters of the reactor and physicochemical properties of gas and liquid phases. The knowledge of the examined in the present work parameters is extremely important to model the operation of a three-phase system in the pulsing flow regime.

Keywords: trickle bed reactors, foaming systems, pulsing flow

### **1. Introduction**

Trickle-bed reactors are commonly used in the branches of industry where great streams of substrates are processed (among others in petrochemical and kerosene industries) (Al-Dahhan et al. (1997), Dudukovic et al. (2002)). Physicochemical properties of gases and liquids, which are either substrates or products in the carried out in TBR processes, differ from air and water properties, that is systems which were applied by many authors in their research (Saroja and Nigam (1996), Duduković et al. (1999, 2002)). Besides, a lot of them (alcohols, solvents whose bases are hydrocarbons, kerosene, diesel fuels) form foams as a result of contact with gas in packing. The occurrence of a foam in the system is not a favourable phenomenon as it

increases gas flow resistance through packing and decreases the amount of liquid present in bed. Thus, it changes the parameters influencing safety and economics of the process unfavourably. Nevertheless, it is a commonly occurring phenomenon in a lot of processes. Thus, it seems advisable to carry out experimental research into the hydrodynamics of the systems.

The aim of the present work was to experimentally determine the parameters characterizing the pulse flow of liquids through bed i.e. the frequency of pulsation and the velocity of pulses travelling along the bed and next to work out the correlation equations enabling their assessment.

## 2. Experimental set-up and measuring system

The research was carried out in a pressure reactor (pressures up to 2 MPa) having 0.05 m in diameter and being 1.5 m long (Fig. 1).

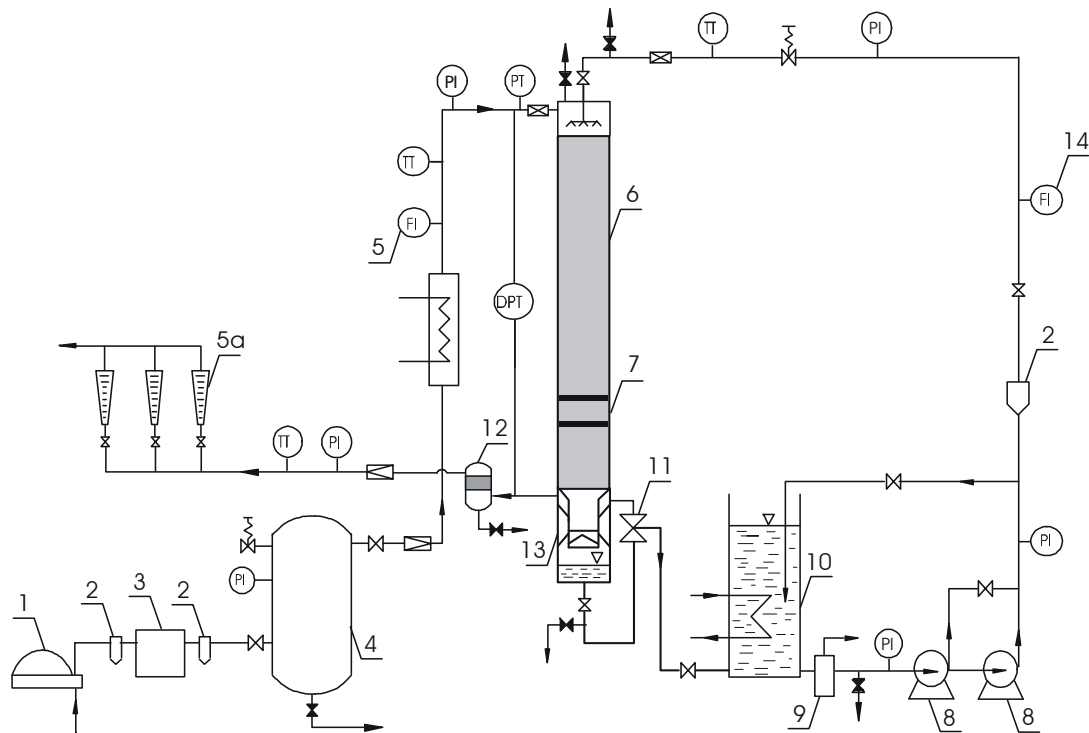


Fig. 1. Experimental set-up: 1 – air compressor, 2 – filters, 3 – air dryer, 4 – compressed air tank, 5 – flowmeters, 6 – packed column, 7 – conductivity cells, 8 – liquid pumps, 9 – microbubble separator, 10 – liquid tank, 11 – float valve, 12 – demister, 13 – gas – liquid separator.

Physicochemical properties of the gas and liquid phases were changed in the research and the concentrations of water solutions of aliphatic alcohols (Table 1) were chosen in such a way so that the systems would form foams of various lifetime as a result of their contact with the gas in packing (WF – weakly foaming, SF – strongly foaming). The method of measuring the changes in conductivity of the two-phase gas-liquid mixture flowing through bed was used to determine the parameters characterizing the pulsing flow of fluids.

Gas phase			Liquid phase				
Medium	$\rho$ [kg·m <sup>-3</sup> ]	$\mu 10^5$ [Pa·s]	Medium (mass percent)	$\rho$ [kg·m <sup>-3</sup> ]	$\mu 10^3$ [Pa·s]	$\sigma_L 10^3$ [N·m <sup>-1</sup> ]	System
air	1.205	1.805	methanol (~53%)	913	1.73	33.20	WF
			methanol (~24%)	960	1.71	44.76	SF
			ethanol (~50%)	914	2.66	29.78	SF
			ethanol (~40%)	935	2.60	31.88	SF
			isopropanol (~56%)	891	3.75	23.90	WF
			isopropanol (~34%)	944	3.41	27.63	SF

Table 1. Physicochemical properties of the tested system ( $t=20^\circ\text{C}$ ), ( $P_r=0.1$  MPa)

It was stated that, for the tested systems, pulse flow regime is obtained at air pressure in the reactor not exceeding 2 MPa for methanol (WF, SF) and ethanol (SF) solutions, but for 56% isopropanol (WF) only up to 0.6 MPa. The region of pulsing flow regime occurrence for pressures higher than atmospheric was not found for 34% isopropanol solution (Janecki et al. (2005)).

### 3. Parameters characterizing pulse fluid flow

To model pulse flow is necessary to know the parameters characterizing it which are the pulsing flow of fluids, namely, the pulse velocity and the frequency of pulsation (Rao and Drinkenburg (1985), Dimenstein and Ng (1986), Dankworth et al. (1990), Bartelmus (1993), Belhouwer i wsp. (2002)).

Only a few works whose authors estimated parameters characterizing pulse fluid flow can be found in literature. In their works the research was carried out in reactors operating at pressure close to atmospheric and the tested systems were, first of all, water – nitrogen or water – air (Weekman and Myers (1964), Sato et al. (1973), Blok and Drinkenburg (1982), Rao and Drinkenburg (1983), Tsochatzidis and Karabelas (1995)). It was only in the works of Bartelmus et al. (1998), Burghardt et al. (1999, 2003a,b), Szlemp et al. (2001) and Tsochatzidis et al. (1998) that an attempt was undertaken at quantification of the dependencies of the measured parameters (that is  $f_p$  and  $V_p$ ) on the reactor operational parameters and physicochemical properties of the liquid spraying the bed (glycerol solutions).

#### 3.1. Velocity of the pulses

The methods enabling tracing the changes in the two-phase structure of the gas-liquid mixture flowing through bed in portions “rich in liquid” and “rich in gas” are used to measure the velocity at which pulses move along the bed. Therefore, they are optical methods (Weekman and Myers (1964), Sato et al. (1973)), acoustic methods (Kolb et al. (1990), Christensen et al. (1986)) or conductometric methods (Blok et al. (1982, 1983), Rao and Drinkenburg (1983), Bartelmus et al. (1998), Burghardt et al. (1999)). In the present work the velocity of pulses was calculated using the signal produced by the two conductometric cells placed in the bed at distances of 1.0 and 1.1m from the top.

The signals from the cells, after amplifying, were fed into the computer memory and collected in 90-second series. Sampling frequency was fixed at the level of 100 Hz. The registered in the computer memory signals were standardized to the form of zero mean value and unitary variance. Having standardized data sequences from two conductometric cells at the distance of  $\Delta l=0.1$  from each other a graph of a cross-correlation function of both signals was plotted. The correlogram showed a distinct maximum (Fig. 2) and the time corresponding to this maximum (termed the signal delay time,  $\tau_{max}$ ) is the time interval required by a pulse to cover the distance between the upper and lower conductometric cells.

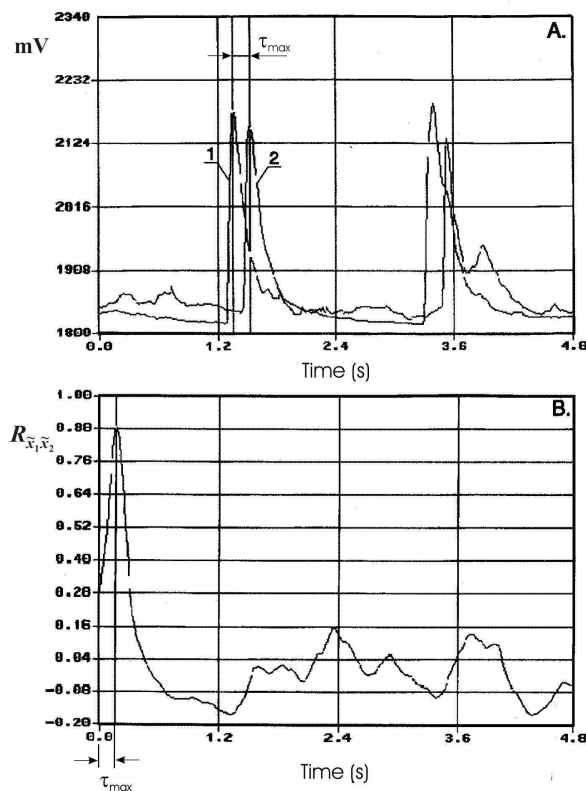


Fig. 2. Graph of the function of signal intercorrelation of signals from two conductometric cells.

The experimentally obtained  $V_p$  values depending on the real velocities of both phases are shown as an example in Fig. 3.

It was stated that pulse velocity was lower than gas real velocity in case of every tested system. It means that gas always penetrates pulse in the systems with low value of surface tension. What follows from the comparison of  $V_p$  values for weakly and strongly foaming systems is that this feature of the system does not influence the velocity of the arising pulses. One has to remember, however, that strongly foaming systems obtain the pulse flow regime at much lower flow velocities of both phases.

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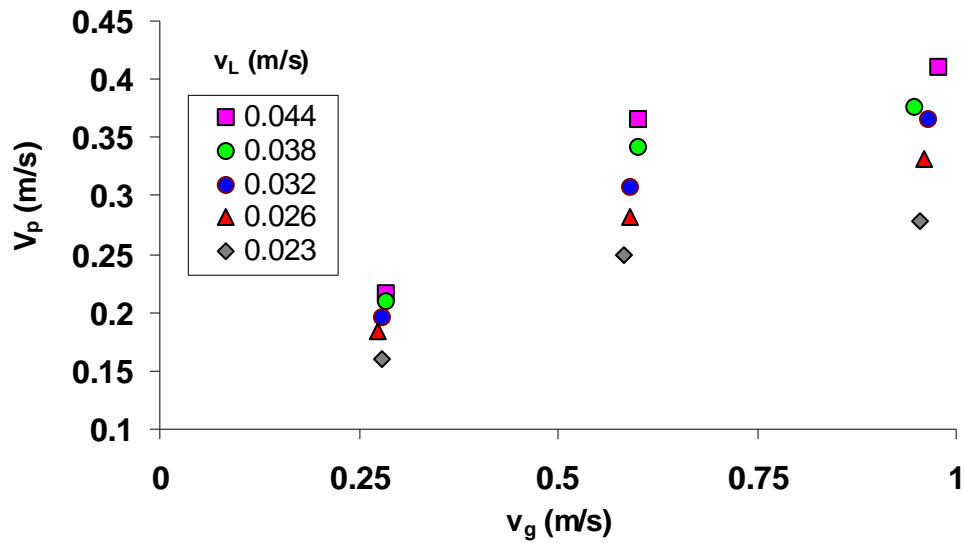


Fig. 3. The dependence of pulse velocity on the real velocities of both phases; isopropanol 56%-air system;  $P=0.6$  MPa.

The change in the density of gas phase (pressure) in the reactor did not influence the measured value significantly (Fig. 4).

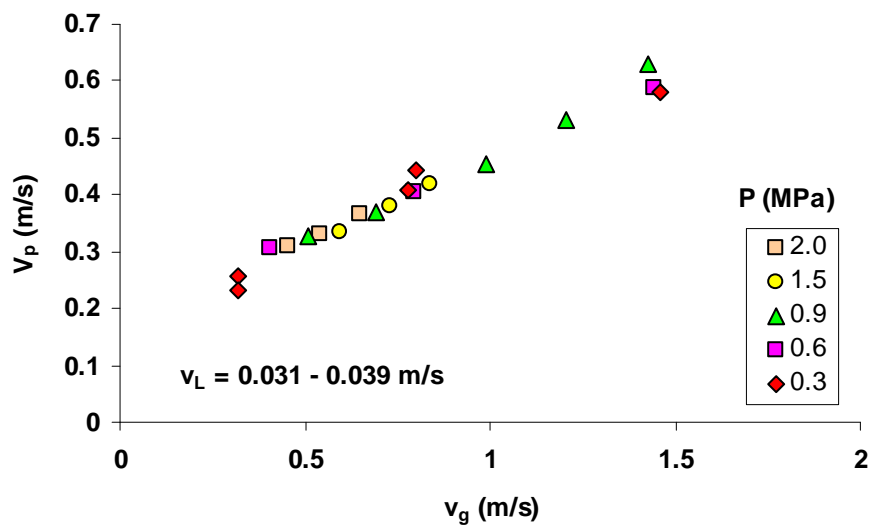


Fig. 4. The influence of gas density on pulsation frequency; methanol 53%-air system.

The data base obtained in the pressurized reactor and in the apparatus working at atmospheric pressure (Burghardt et al. (2003a, 2003b)) makes it possible to elaborate an equation correlating the measured values of pulsation velocity. The following dependence was obtained:

$$\frac{V_p}{v_g} = \left[ 1.6818 Re_{gr}^{-0.2857} - 0.213 \left( \frac{\rho_{airN}}{\rho_g} \right)^{0.406} \right] Re_{Lr}^{0.358} \quad (1)$$

which with the mean percentage error of 10.5% describes the measured  $V_p$  values ( $R=0.957$ ) (Fig. 5).

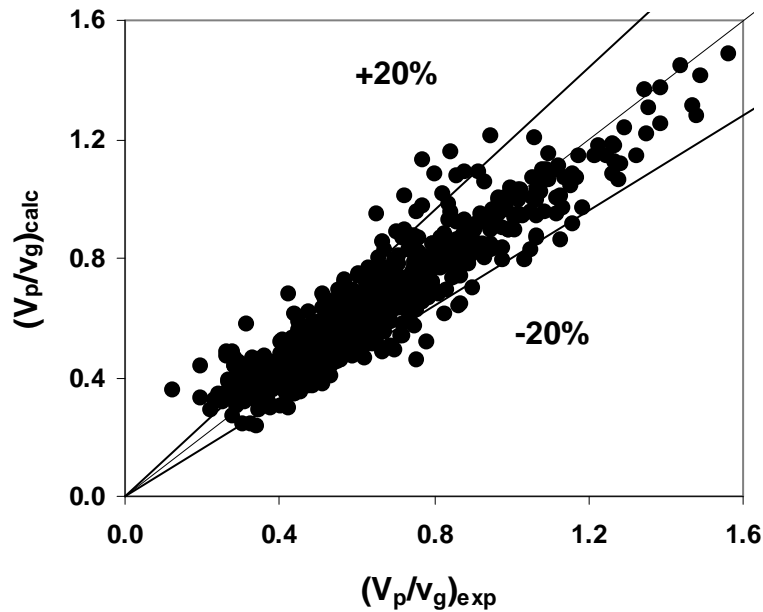


Fig. 5. The comparison of  $V_p$  values estimated experimentally and calculated from the equation (1)

### 3.2. Pulsation frequency

Pulsation frequency was determined by analysing the signal from the upper conductometric cell placed in bed  $\sim 1.0$ m below the top of bed. The procedure described in detail in the work of Burghardt et al. (1999) was used to obtain the aim. It enables obtaining a set of pulse duration and the number of pulses with the given duration for every 90-second realization. Next, probability density function was calculated from the test and its chart (histogram) was drawn. A log-normal distribution was accepted as a statistical model as a result of the analysis of its shape. A mean pulse duration was determined from the first moment of this distribution, whose converse is the looked for value of pulsing frequency.

While analysing the obtained in the present work findings of the experiments for systems forming foams it was stated that it is very difficult to define the influence of real velocities of both phases on the pulse frequency as this dependence is different for systems forming weakly and strongly foams (Fig. 6).

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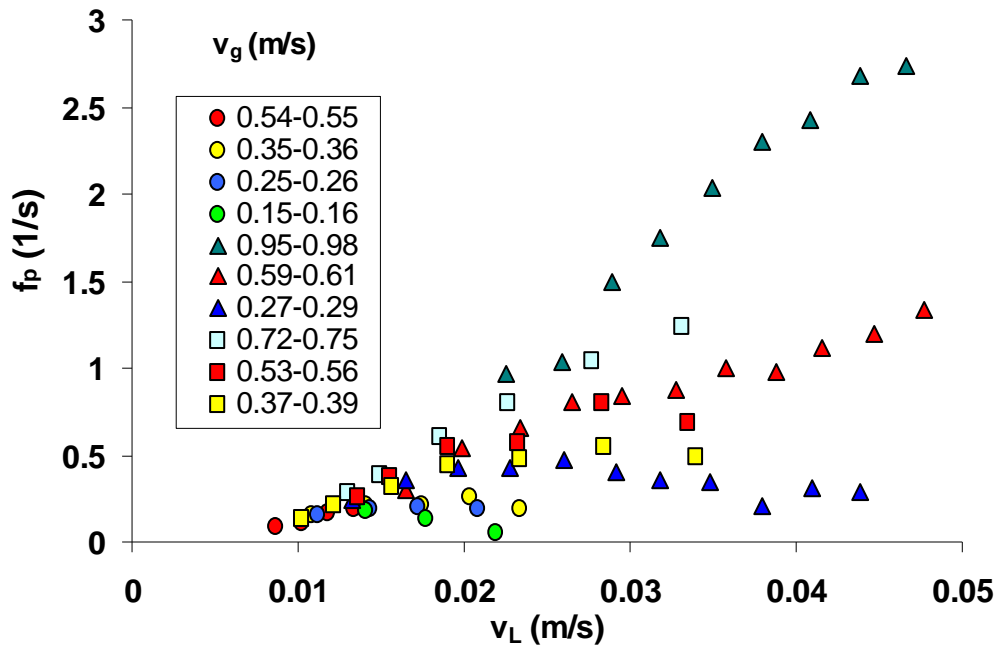


Fig. 6. The influence of real velocities of both phases on the pulsation frequency; (●) ethanol 50%-air system, (■) ethanol 40%-air system, (▲) isopropanol 56%-air system,  $P=0.6$  MPa.

As well as this, the change in gas/pressure density in the reactor causes the change in the number of pulses formed in the column; the higher the gas density in the reactor, the higher the pulsation frequency (Fig. 7).

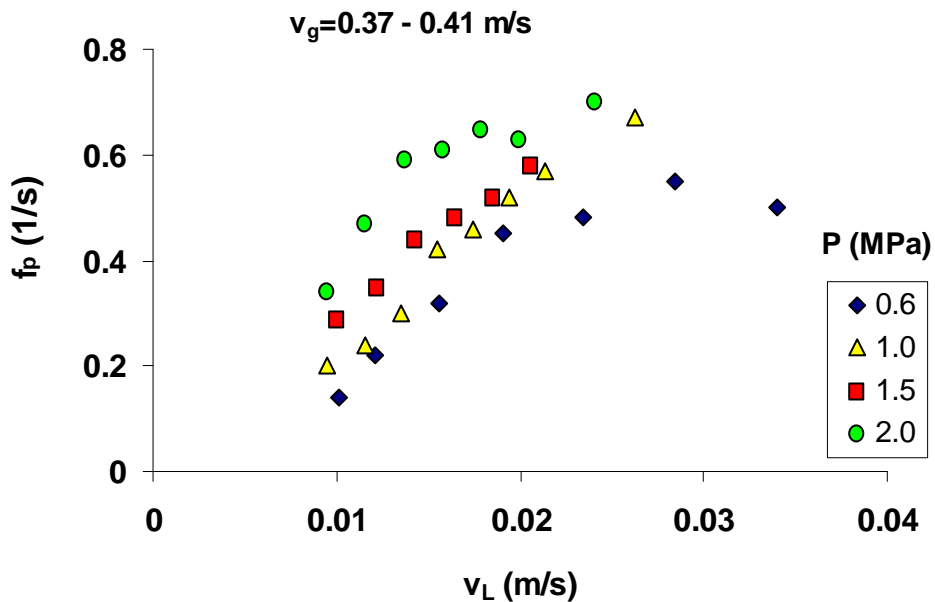


Fig. 7. The influence of gas density on pulsation frequency; ethanol 40%-air system.

The  $\psi$  flow parameter was introduced into the correlation equation to catch hold of influence of the physicochemical properties of the tested solutions on the measured  $f_p$  values. The following equations have been proposed:

- for strongly foaming systems (234 experimental points):

$$f_p = 0.2354 + 0.63\psi^{1.2916} Re_{gr}^{0.2697} We_{Lr} \quad (2)$$

which with the mean percentage error where  $e_Y=33.4\%$  ( $R=0.936$ ) approximates the experimental data.

- for weakly foaming systems (387 experimental points):

$$f_p = 0.343 Re_{gr}^{0.1655} + 0.0062\psi^{1.22} Re_{gr}^{0.447} We_{Lr} \quad (3)$$

where  $e_Y=28.3\%$  ( $R=0.824$ ).

The region of low pulsation frequency is the most interesting one for industrial purposes because only then, from the economic point of view, the increase in resistance of gas flow through packing is compensated for by a great increase in mass transfer coefficients. In the initial phase of pulsation the frequency increases in a linear way together with the increase in the real velocity of the liquid phase. For a reactor operating at an elevated pressure the majority of experimental points was located in the region of linear dependence of the pulsation frequency on  $v_L$ .

In addition, it was noticed that the directivity factor of these straight lines changes both with the change in gas phase velocity and pressure in the reactor. However, changes in the physicochemical properties of liquid are take into account in the real velocity of the liquid phase.

In the considered region the following dependence was obtained for weakly foaming systems (200 experimental points):

$$f_p = (36.629 + 1.211 Re_{gr}^{0.406}) v_L - 0.4212 \quad (4)$$

which with the mean relative error not exceeding 18% ( $R=0.937$ ) approximates the experimental data (Fig. 8).

#### 4. Summary

The results of the research whose aim was to determine parameters characterizing pulsing flow of fluids in a trickle-bed (gas-liquid-solid) reactor with a fixed bed were presented in the work. The research was carried out for a specific group of measuring systems – systems which formed foams in the pulse flow regime. The aim of the experiments was to find the answer to the question how the change in physicochemical properties of fluids and the forming of foams in the system influence the value of the measured parameters.



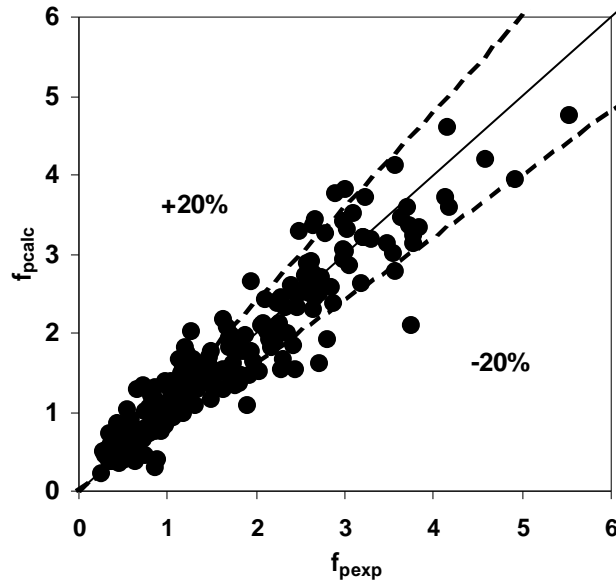


Fig. 8. The comparison of  $f_p$  values experimentally determined and calculated from equation (4)

The change of the hydrodynamic regime of the apparatus to pulse flow regime and next calculating the productivity and selectivity of the process requires the knowledge of parameters characterizing this hydrodynamic regime (the velocity of pulses and their frequency). The carried out in the present work research and the correlations formulated on its basis make it possible to calculate the values of these parameters and as for system forming foams they have been the only available dependencies so far.

#### Notation

$d_p$  – particle diameter (m)

$$e_Y = \frac{1}{N} \sum_{i=1}^N \left| \frac{Y_{exp,i} - Y_{cal,i}}{Y_{exp,i}} \right| \cdot 100\% \quad \text{– mean percentage error (\%)}$$

$f_p$  – frequency of pulsation ( $s^{-1}$ )

$P$  – pressure (Pa)

$R$  – correlation coefficient

$R_{\tilde{x}_1, \tilde{x}_2}$  – cross-correlation function

$v_\alpha$  – real velocity of  $\alpha$  phase ( $ms^{-1}$ )

$V_p$  – pulse velocity (m)

#### Greek letters

$\mu$  – viscosity (Pas)

$\rho$  – density ( $kg\ m^{-3}$ )

$\tau$  – time (s)

$\sigma$  – surface tension ( $N\ m^{-1}$ )

*Dimensionless numbers*

$$Re_{\alpha r} = \frac{v_{\alpha} \cdot d_p \cdot \rho_{\alpha}}{\mu_{\alpha}} \quad - \text{ Reynolds number}$$

$$We_{Lr} = \frac{v_L^2 \cdot \rho_L \cdot d_p}{\sigma_L} \quad - \text{ Weber number}$$

$$\psi_L = \frac{\sigma_w}{\sigma_L} \left[ \frac{\mu_L}{\mu_w} \left( \frac{\rho_w}{\rho_L} \right)^2 \right]^{0.33} \quad - \text{ flow parameter}$$

*Subscripts*

- g     – gas
- L     – liquid
- max   – maximum
- N     – normal conditions
- p     – pulse
- w     – water
- $\alpha$    –  $\alpha$ -phase,  $\alpha = g, L$

**References**

- Al-Dahhan M.H., Larachi F., Duduković M.P., Laurent A., (1997) *Industrial Engineering Chemistry Research*, 36, 3292-3314.
- Bartelmus G., (1993) *Chemical Engineering and Processing*, 32, 65-74.
- Bartelmus G., Gancarczyk A., Stasiak M., (1998) *Chemical Engineering and Processing*, 37, 331-341.
- Boelhouwer J.G., Piepers A.A., Drinkenburg A.A.H., (2002) *Chemical Engineering Science*, 57, 3387-3399.
- Blok J.R., Drinkenburg A.A.H., (1982) *Chemical Engineering Journal*, 25, 89-99.
- Blok J.R., Varkevisser J., Drinkenburg A.A.H., (1982) *Chemical Engineering Journal*, 25, 89-99.
- Blok J.R., Varkevisser J., Drinkenburg A.A.H., (1983) *Chemical Engineering Science*, 38, 687-699.
- Burghardt A., Bartelmus G., Gancarczyk A., (1999) *Chemical Engineering and Processing*, 38, 411-426.
- Burghardt A., Bartelmus G., Janecki D., (2003a) *Chemical and Process Engineering*, 24, 151-164.
- Burghardt A., Bartelmus G., Janecki D., (2003b) *Chemical and Process Engineering*, 24, 165-182.
- Christensen G., McGovern S.J., Sundaresan S., (1986) *AIChE Journal*, 32, 1677-1689.
- Dankworth D.C., Kevrekidis I.G., Sundaresan S., (1990) *AIChE Journal*, 36, 605-621.
- Dimenstein D. M., Ng K. M., (1986) *Chemical Engineering Communications*, 41, 215-235.

- Dudukovic M.P., Larachi F., Mills P.L., (2002) *Catalysis Review*, 44, 123-246.
- Dudukovic, M. P., Larachi F., Mills P. L., (1999) *Chemical Engineering Science*, 54, 1975-1995.
- Janecki D., Bartelmus G., Krótki T., 7th World Congress on Chemical Engineering Proceedings, 10-14 July 2005, Glasgow, Scotland.
- Kolb W.B., Melli T.R., de Santos M.J., Scirven L.E., (1990) *Industrial Engineering Chemistry Research*, 29, 2380-2389.
- Rao V.G., Drinkenburg A.A.H., (1983) *Canadian Journal of Chemical Engineering*, 61, 158-167.
- Rao V.G., Drinkenburg A.A.H., (1985) *AIChE Journal*, 31, 1010-1018.
- Sato Y., Hirose T., Takahashi F., Toda M., Hashiguchi Y., (1973) *Journal Chemical Engineering of Japan*, 6, 315-319.
- Sorosha A.K., Nigam K.D.P., (1996) *Review Chemical Engineering* 12, Nos. 3-4.
- Szlemp A., Bartelmus G., Janecki D.; (2001) *Chemical Engineering Science*, 56, 1111-1116.
- Tsochatzidis N.A., Karabelas A.J., (1995) *AIChE Journal*, 41, 2371-2382.
- Tsochatzidis N.A., Ntampeglitis K.J., Karabelas A.J., (1998) *Chemical Engineering Communications*, 166, 137-156.
- Weekman V.W., Myers J.E., (1964) *AIChE Journal*, 10, 951-957.