

PRESSURE DROP IN TWO-PHASE GAS-LIQUID (TAYLOR) FLOW IN MICROREACTORS

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The pressure drop in microreactors for the gas – liquid Taylor flow was measured for 4 different microreactor geometries and 3 different gas – liquid systems. The results have been compared with the existing literature correlations. A selection of the best correlations has been made.

Keywords: gas - liquid Taylor flow, pressure drop, microreactor

1. INTRODUCTION

Pressure drop in two-phase gas-liquid flow in pipes and channels has been the subject of research for many decades. Most experimental data existing in the literature have been obtained for a conventional industrial equipment (Dziubiński and Prywer, 2009).

Recently, microreactors and, more generally, microstructures, have become increasingly important parts of modern process equipment. It is important to know, whether the correlations obtained for industrial size channels are applicable to milli- and microchannels. Three approaches to calculate pressure drop values in such channels have been used: the classical approach (Fries et al., 2008), the homogenous model (Kawahara et al., 2002) and the Lockhart-Martinelli (1949) model. Tab. 1 shows the existing information about the pressure drop investigations in micro and milli-channels.

The aim of this paper is to report experimental results obtained for three different systems, and a number of channel geometries and positions (horizontal, vertical). The obtained data have been correlated using the aforementioned three models.

The total pressure drop in gas-liquid microchannels includes four different contributions (Fries et al., 2008):

$$\Delta P_T = \Delta P_A + \Delta P_B + \Delta P_G + \Delta P_F \quad (1)$$

where:

ΔP_A is the pressure drop due to acceleration (Fries et al., 2008):

$$\Delta P_A = G^2 \left[\left(\frac{x^2}{\beta \cdot \rho_G} + \frac{(1-x)^2}{(1-\beta) \cdot \rho_L} \right)_{outlet} - \left(\frac{x^2}{\beta \cdot \rho_G} + \frac{(1-x)^2}{(1-\beta) \cdot \rho_L} \right)_{inlet} \right] \quad (2)$$

ΔP_B is the pressure drop over a moving bubble due to a different shape of the bubble front and bubble tail (Bretherton, 1961):

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$$\Delta P_B = 3.58 \cdot \frac{\sigma}{r} \cdot \sqrt[3]{9 \cdot Ca^2} \quad (3)$$

ΔP_G is the pressure drop due to gravity (Fries et al., 2008):

$$\frac{\Delta P_G}{L} = (\beta \cdot \rho_G + (1 - \beta) \cdot \rho_L) \cdot g \cdot \sin(\alpha) \quad (4)$$

ΔP_F is the frictional pressure drop, which can be determined in a number of ways:

- according to the classical approach using the Darcy equation for liquid flow correlated for the fact that only a fraction of the channel volume is filled by the liquid:

$$\frac{\Delta P_F}{L} = f \cdot \rho_L \cdot u_{TP}^2 \cdot \frac{2}{d} \cdot \varepsilon_L \quad (5)$$

- according to the homogenous model using the Darcy equation for the two-phase mixture:

$$\frac{\Delta P_F}{L} = f_H \cdot \rho_{TP} \cdot u_{TP}^2 \cdot \frac{2}{d} \quad (6)$$

- according to the Lockhart-Martinelli model, as discussed in the following section.

Table 1. Pressure drop investigations in micro and milli-channels

	Channel cross-section	System	Flow direction	Dimensions of main channel (wide x depth x length or diameter x length) [mm]	Hydraulic diameter [mm]	Channel length to hydraulic diameter ratio	Model
Bretherton, 1961	circular	air and water	horizontal	5x1000	5	200	classical
Chisholm, 1967	circular	air and water	no data available	no data available	1÷250	no data available	Lockhart-Martinelli
Mishima and Hibiki, 1996	circular	air and water	horizontal	1.05x210; 2.05x310; 3.12x510; 4.08x1000	1.05; 2.05; 3.12; 4.08	200; 151; 163; 245	Lockhart-Martinelli
Lee and Lee, 2001	rectangular	air and water	vertical	0.4x20x640; 1x20x640; 2x20x640; 4x20x640	0.78; 1.91; 3.64; 6.67	499; 335; 175; 95	Lockhart-Martinelli
Kawahara et al., 2002	circular	air and water	vertical	0.1x64.5	0.1	645	homogenous, Lockhart-Martinelli
Yue et al., 2004	rectangular square	nitrogen and water	horizontal	0.575x0.488x34.30 0.333x0.332x34.52	0.528 0.333	65.0 103.7	homogenous, Lockhart-Martinelli
Kreutzer et al., 2005a	circular	air and water/decane/tetradecane	horizontal	no data available	2.3	no data available	classical
Saisorn and Wongwises, 2010	circular	air and water	vertical	0.15x104; 0.22x120; 0.53x320	0.15; 0.22; 0.53	693; 545; 603	Lockhart-Martinelli
The present paper	circular square	nitrogen and water/ethanol/heptane	vertical and horizontal	0.15x25; 0.4x1000	0.15; 0.4	166; 250	classical, homogenous, Lockhart-Martinelli

The pressure drop contributions defined by Eqs. (2-4) are simple to calculate from the experimental conditions.

It should be pointed out that all traditional models (classical, homogenous and Lockhart-Martinelli) are based on the assumptions which in the case of the Taylor flow are rather unrealistic. A more rational approach has been suggested by Kreutzer and co-workers (2005a, 2005b, 2005c) and Abiev (2011). However, their approach requires the knowledge of slug length, which renders it less practical from the engineering point of view. For this reason we limited our analysis to the traditional models.

1.1. The classical model (Fries et al., 2008)

In this model the Darcy Equation (5) is used, with the physicochemical properties (ρ_L and μ_L) of the liquid, and two-phase superficial velocity u_{TP} . The friction factor f is given by Equations (7-9).

Standard correlation:
$$f = \frac{16}{Re} \quad (7)$$

Bretherton (1961):
$$f = \frac{16}{Re} \left[1 + 0.465 \cdot \frac{d_H}{L_L} \cdot \left(\frac{1}{Ca} \right)^{\frac{1}{3}} \right] \quad (8)$$

Kreutzer et al. (2005a):
$$f = \frac{16}{Re} \left[1 + 0.17 \cdot \frac{d_H}{L_L} \cdot \left(\frac{Re}{Ca} \right)^{\frac{1}{3}} \right] \quad (9)$$

1.2. The homogenous model (Kawahara et al., 2002)

The homogeneous model assumes that the two phases (gas and liquid) form a homogenous mixture. The pressure drop is calculated on the basis of one-phase flow formulas with the appropriate two-phase mixture parameters values. Although this assumption is rather far from the physical reality of the Taylor flow, we decided to check the applicability of this model for the sake of completeness of the analysis.

The literature describes many correlations for the viscosity and density of gas-liquid mixture determination. In this paper the following correlations are discussed:

- density:

Kawahara et al., (2002):
$$\frac{1}{\rho_H} = \frac{x}{\rho_G} + \frac{1-x}{\rho_L} \quad (10)$$

- viscosity:

Owens (1961):
$$\mu_{TP} = \mu_L \quad (11)$$

Dukler et al., (1964):
$$\mu_{TP} = \varepsilon_G \cdot \mu_G + (1 - \varepsilon_G) \cdot \mu_L \quad (12)$$

Beattie and Walley (1982):
$$\mu_{TP} = (1 - \varepsilon_G) \cdot \mu_L \cdot (1 + 2.5\varepsilon_G) + \varepsilon_G \cdot \mu_G \quad (13)$$

Using the calculated values of density and viscosity of the gas-liquid mixture, the frictional pressure drop can be calculated from the Darcy equation in the form given in Eq. (6).

The frictional factor is determined from the correlation (14):

$$f_H = \frac{16}{\text{Re}_H} \quad (14)$$

1.3. The Lockhart and Martinelli model

Lockhart and Martinelli (1949) proposed that the pressure drop in two-phase flow can be related to the equivalent pressure drop in one-phase flow by two-phase parameter Φ :

$$\left(\frac{\Delta P_F}{L}\right)_{TP} = \phi_L^2 \cdot \left(\frac{\Delta P_F}{L}\right)_L = \phi_G^2 \cdot \left(\frac{\Delta P_F}{L}\right)_G \quad (15)$$

where Φ_L is correlated as (Chisholm, 1967):

$$\phi_L^2 = 1 + \frac{C}{X} + \frac{1}{X^2} \quad (16)$$

and X is the ratio:

$$X^2 = \frac{\left(\frac{\Delta P_F}{L}\right)_L}{\left(\frac{\Delta P_F}{L}\right)_G} \quad (17)$$

The pressure drop for the gas phase and the liquid phase is calculated separately using the Darcy equation for one phase (18) and the standard definition of the friction factor f , using Eq. (7).

$$\frac{\Delta P_F}{L} = f \cdot \rho \cdot u^2 \cdot \frac{2}{d} \quad (18)$$

The main problem in applying the Lockhart-Martinelli model is to determine the C factor. Table 2 shows the existing C factor correlations for laminar gas-liquid flows.

Table 2. C factor correlations

	C correlation	Eq. number
Chisholm (1967)	$C = 5$	(19)
Mishima and Hibiki (1996)	$C = 21 \cdot [1 - e^{(-319 \cdot d)}]$	(20)
Lee and Lee (2001)	$C = 6.833 \cdot 10^{-8} \cdot \lambda^{-1.317} \cdot \text{Re}_L^{0.557} \cdot \text{Ca}^{0.719}$ $\lambda = \frac{\mu_L^2}{\rho_L \sigma d}$	(21)
Saisorn and Wongwises (2010)	$C = 7.599 \cdot 10^{-3} \cdot \lambda^{-0.631} \cdot \text{Re}_L^{-0.008} \cdot \text{Ca}^{0.005}$	(22)

In this work we used our experimental data to find a new C correlation. We also investigated the contribution of inlet effects to the pressure drop in microreactors.

2. EXPERIMENTAL SYSTEM

Two kinds of microreactors were used: a microreactor built of polydimethylsiloxane (PDMS) and another one, built of glass. In order to eliminate inlet and outlet effects, the inlet/outlet systems, without

the main channel, were also prepared. For each set of experimental conditions the pressure drop was measured twice: for the whole microreactor, and for the inlet system only. It allows to calculate pressure drop in microchannel alone .

The glass microreactor has a circular cross-section, 0.4 mm in diameter and 100 mm of the main channel's length. Fig. 1 shows the glass microreactor.



Fig. 1. Glass microreactor

The PDMS microreactors have a square cross-section, 0.15 mm wide and the main channel 25 mm in length. A PDMS microreactor with a Y-shaped inlet is shown in Fig. 2a), a PDMS microreactor with a T-shape inlet is shown in Fig. 2b), and a PDMS microreactor with a T1-shaped inlet is shown in Fig. 2c).

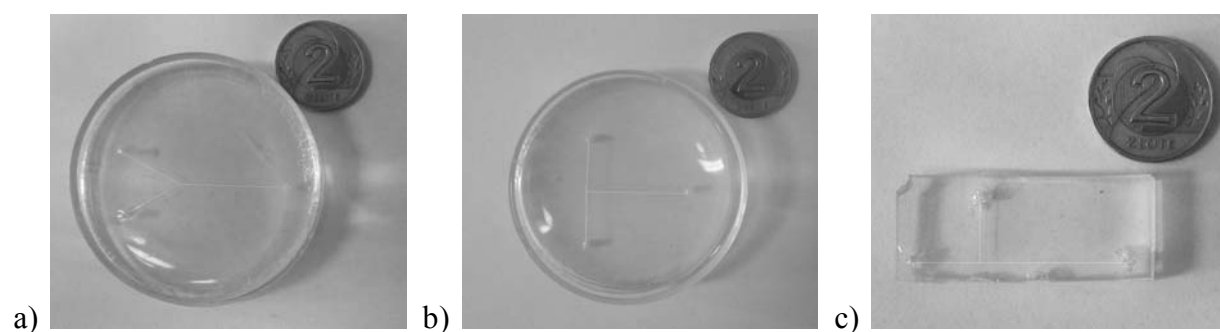


Fig. 2. PDMS microreactors photo: a) Y-shape, b) T-shape, c) T1-shape

In the T-shape microreactor the main channel is perpendicular to both inlet channels, in the T1-shape one of inlet channels is perpendicular to the main channel, while the other lies on its axis. The T1-shape microreactor was used in two different configurations (Fig. 3a) 3b)), T-shape arrangement is shown in Fig. 3c).

The experimental set-up is shown in Fig. 4. Gas (nitrogen) was supplied from a cylinder (1) through a reducing valve and a gas flow regulator (2) (mass flow meter and controller: Brooks Smart Series, type 5850 S). Gas pressure was measured using manometer (5). The liquid was supplied by a syringe pump (3). At the outlet of the microreactor (4) the atmospheric pressure was maintained. In the experiments three liquids were used (water, ethanol and heptane).

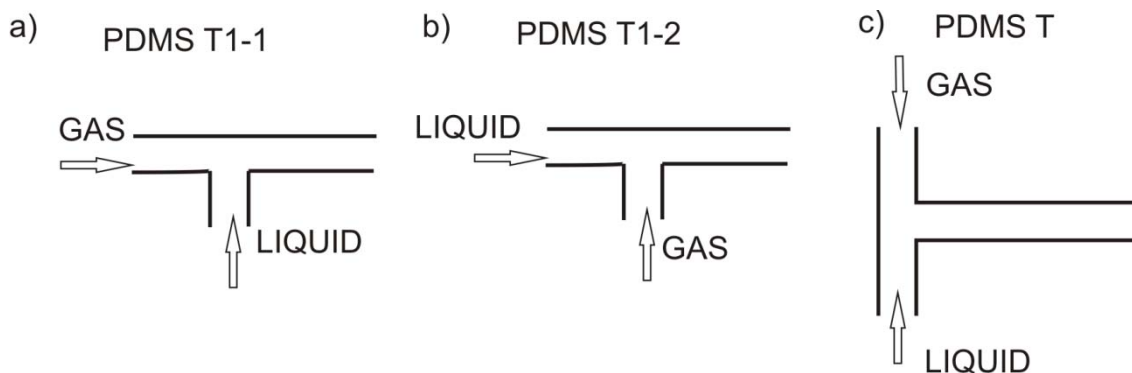


Fig. 3. Scheme of the gas and liquid inlets in PDMS microreactors:

- a) PDMS T1-shape, liquid in perpendicular channel (denoted as PDMS T1-1), b) PDMS T1-shape, gas in perpendicular channel (denoted as PDMS T1-2), c) PDMS T-shape

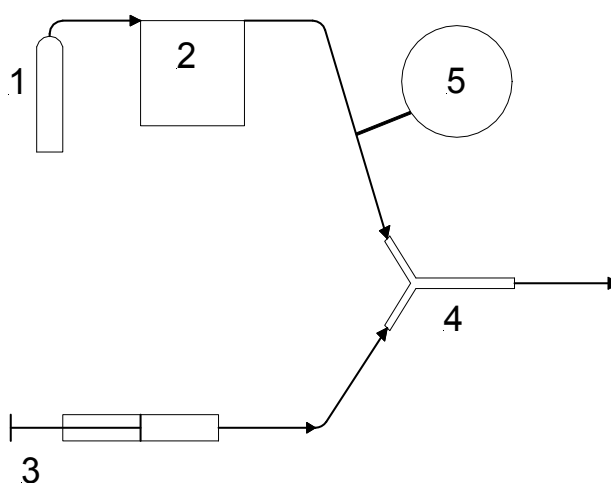


Fig. 4. The experimental set-up:

1-gas cylinder, 2-gas flow regulator, 3-syringe pump, 4-microreactor or inlet system, 5-manometer

3. RESULTS AND DISCUSSION

3.1. Influence of microreactor position on pressure drop

The aim of this part of the experiment was to check whether the position (horizontal or vertical) of the main channel had any influence (other than hydrostatic pressure) on the overall pressure drop. It was supposed that this position may influence the process of bubble/slug formation and/or the character of the liquid flow around the bubble.

The pressure drop was measured for two positions of microreactors: with main microchannel oriented horizontally or vertically. Fig. 5 shows the overall pressure drop ΔP_{Σ} in glass microreactor in systems nitrogen-heptane and nitrogen-water for vertical and horizontal positions.

We also measured the overall pressure drop ΔP_{Σ} in two position (horizontal and vertical) in PDMS-T and PDMS-T1 microreactors in nitrogen-ethanol system (see Fig. 6).

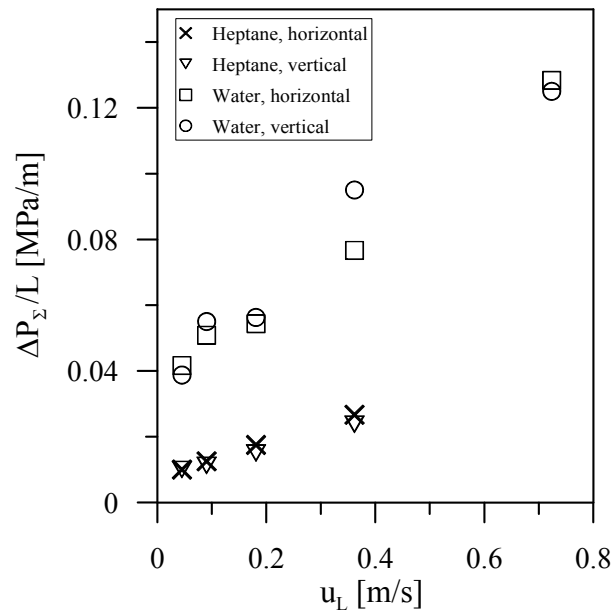


Fig. 5. The influence of microreactor (glass) position on overall pressure drop ($u_G=0.1$ m/s)

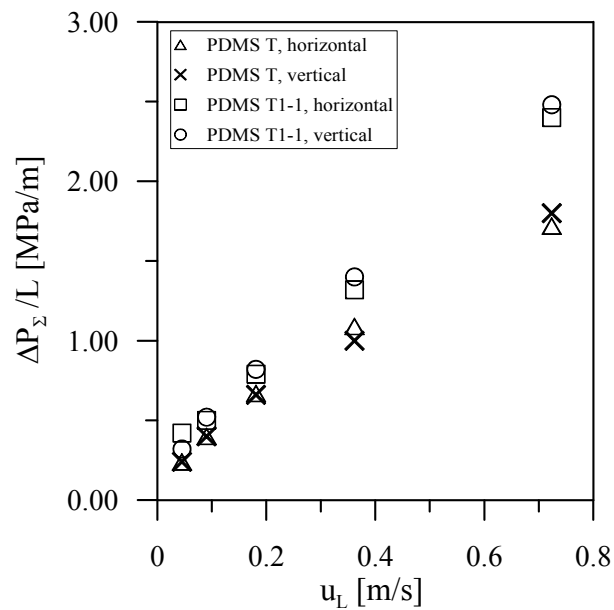


Fig. 6. The influence of microreactor (PDMS) position on overall gradient pressure drop ($u_G=0.7$ m/s)

As can be seen, for lower values of liquid velocity the overall influence of microreactor position on the pressure drop is not significant (the channel is quite short, so the hydrostatic component, Eq. (4), is not important). For higher velocity values the influence of microchannels position on overall pressure drop is not higher than 10%. In further considerations only horizontal microreactors were discussed.

3.2. Inlet effects

Fig. 7 shows the results, obtained using the glass microreactor. Ethanol, water and heptane were used to measure pressure drop in this microreactor and in the inlet system. Fig. 8 shows the experimental data for ethanol-nitrogen system for PDMD T and PDMS T1-1 microreactors and the inlet systems.

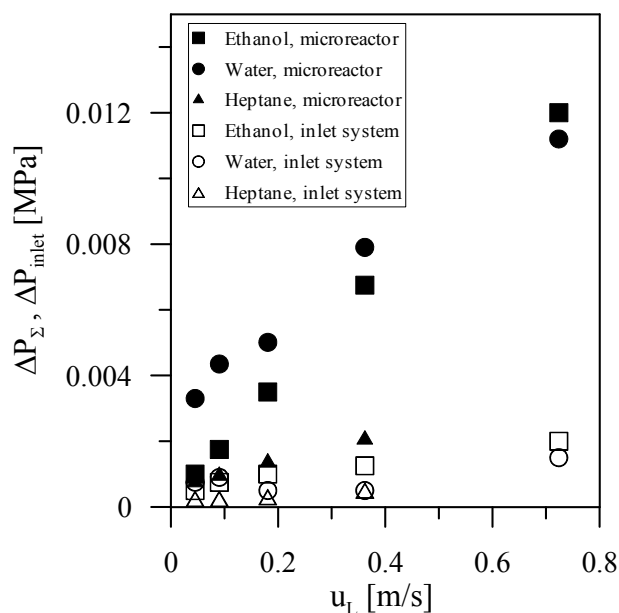


Fig. 7. Comparison of the pressure drop in the microreactor and the inlet system (glass microreactor, ethanol, water, heptane). Superficial gas velocity is constant ($u_G = 0.1$ m/s)

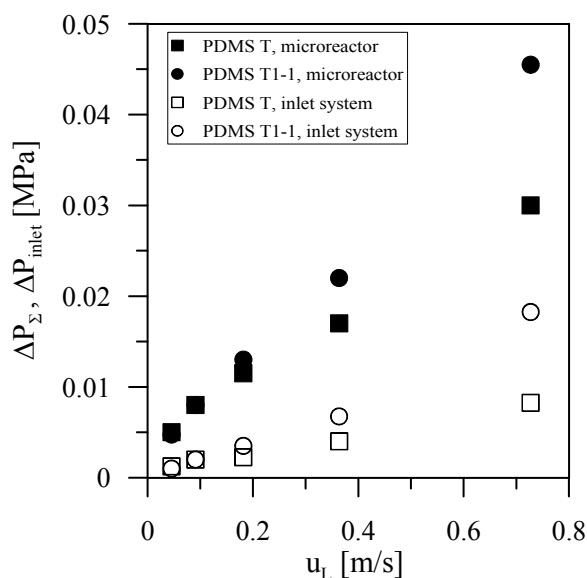


Fig. 8. Comparison of the pressure drop in microreactors and inlet systems (PDMS T and PDMS T1-1, ethanol). Superficial gas velocity is constant ($u_G = 0.1$ m/s)

As can be seen the contribution of the inlet pressure drop in the microreactors was in the range: 10% ÷ 40% of the total measured pressure drop. It is therefore essential to distinguish between the two contributions: that of the inlet system and that of the main channel. Naturally, for longer main channels, the inlet contribution becomes less important.

3.3. Microchannel pressure drop

According to Eq. (1) the total pressure drop in gas-liquid flow in a microchannel is composed of four parts. The frictional pressure drop constitutes the main contribution in all our data. Fig. 9 shows results for the glass microreactor, Fig. 10 shows results for the PDMS microreactor.

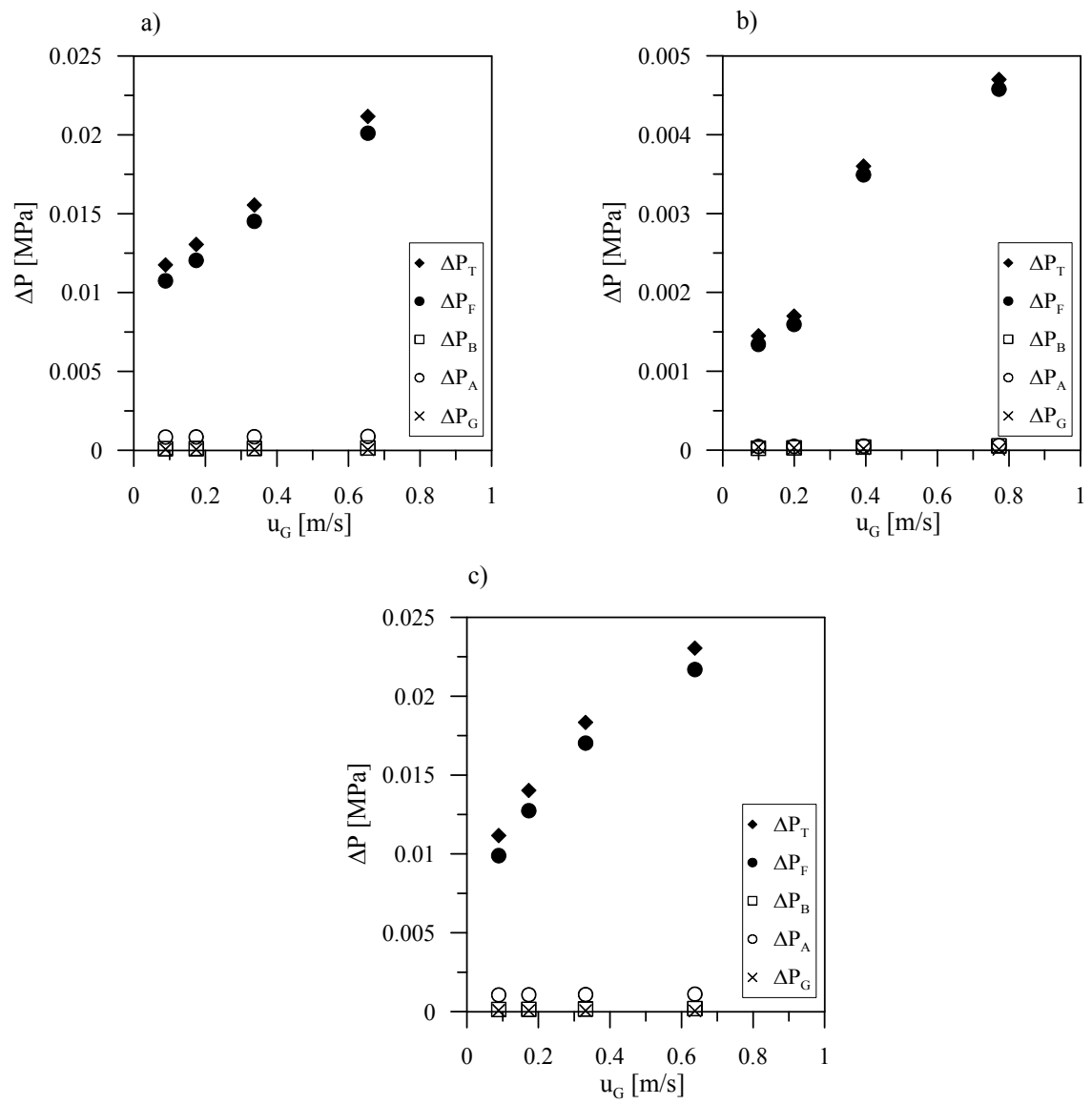


Fig. 9. Contributions to the total pressure drop in the glass microchannel: a) ethanol, $u_L=0.73$ m/s, b) heptane, $u_L=0.18$ m/s, c) water, $u_L=0.73$ m/s

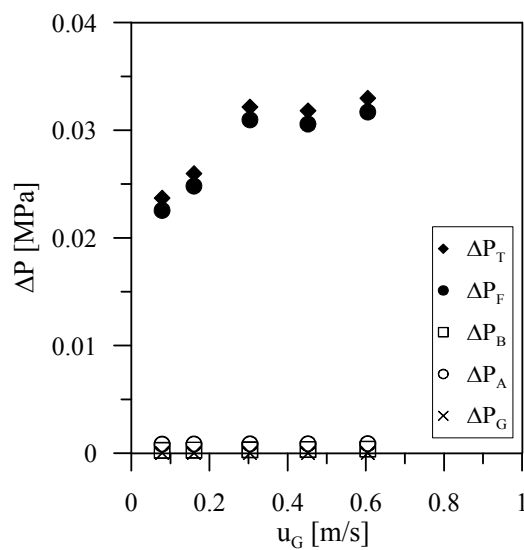


Fig. 10. Contribution to the total pressure drops in the PDMS microchannel for ethanol, and $u_L = 0.73$ m/s

As can be seen the total pressure drop in the microchannel is practically equal to the frictional pressure drop. Our experimental data have been compared with all the three models discussed earlier.

3.4. The classical model

For the classical model a comparison of the experimental data with those calculated from Eq. (5) using three correlations for the factor f Eqs. (7-9) has been made. A comparison of the calculated frictional pressure drop values with the experimental data is shown in Fig. 11. For a quantitative comparison the standard deviation values were calculated as:

$$\sigma_s = \frac{\sum_{i=1}^N \sqrt{\left(\left(\frac{\Delta P_F}{L} \right)_{i(\text{experimental})} - \left(\frac{\Delta P_F}{L} \right)_{i(\text{correlation})} \right)^2}}{N} \quad (23)$$

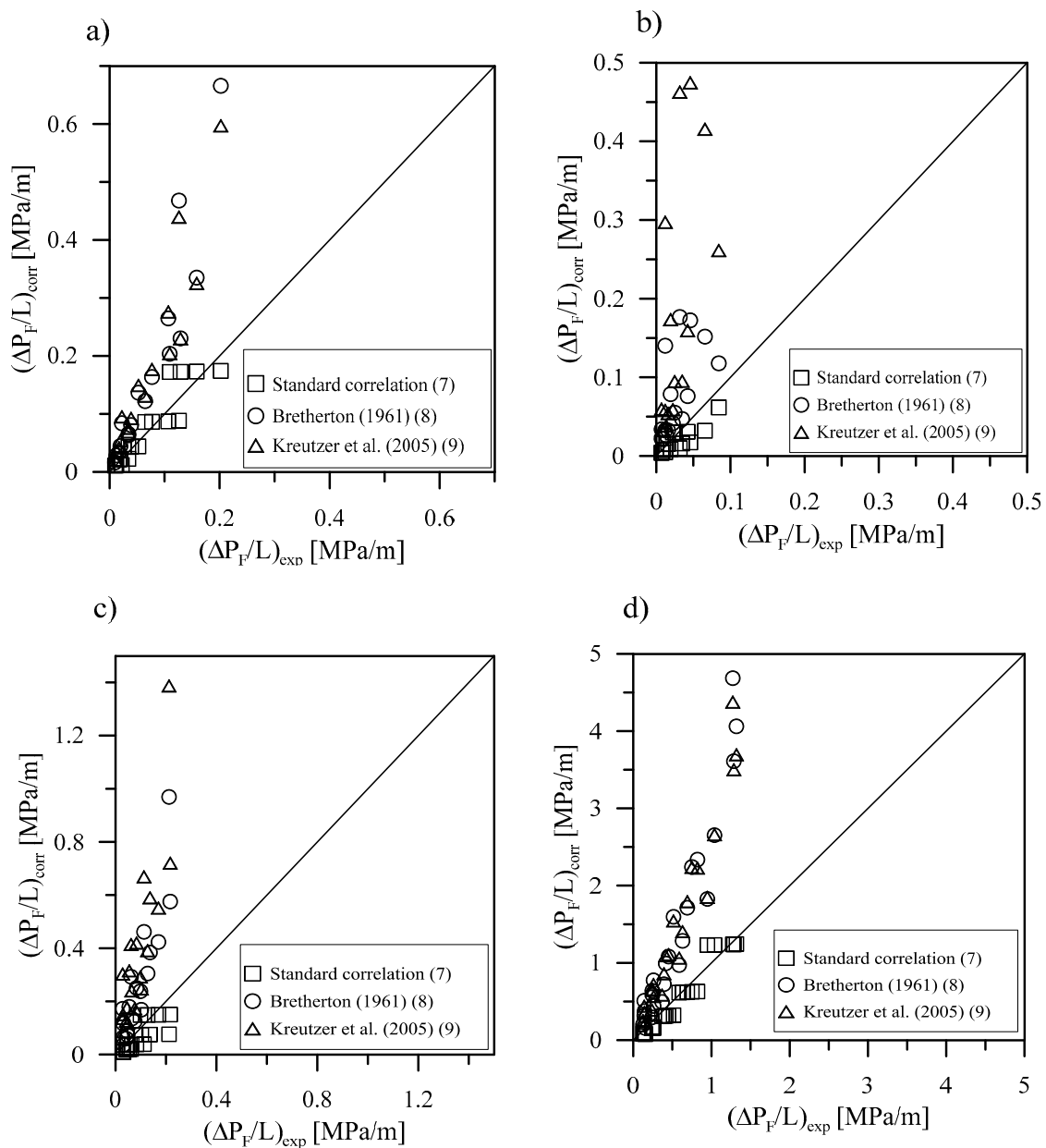


Fig. 11. Comparison of the experimental data with the existing correlations (7-9) for the frictional coefficient: a) glass microreactor, water-nitrogen; b) glass microreactor, heptane-nitrogen system; c) glass microreactor, water-nitrogen; d) PDMS microreactor, ethanol-nitrogen

The calculated values of the standard deviation are shown in Table 3

Table 3. The values of standard deviation for the considered frictional coefficient correlations in the classical model (MPa/m)

system ↓	f →	Classical model		
		Bretherton (1961), Eqs. (5, 8)	Standard correlation, Eqs. (5, 7)	Kreutzer et al. (2005a), Eqs. (5, 9)
Glass Ethanol-nitrogen		0.157	0.023	0.147
Glass Heptane-nitrogen		0.073	0.016	0.106
Glass Water-nitrogen		0.231	0.048	0.382
PDMS Ethanol-nitrogen		1.196	0.112	1.150
Average		0.414	0.049	0.446

It is seen, that the classic model with the standard correlation for f (Eqs. 5, 7) gives a relatively better agreement than the Bretherton (Eq. 8) and the Kreutzer (Eq. 9) correlations. The standard deviation values are in the range from 0.016 to 0.112 MPa/m. It should be pointed out that in the case of Bretherton and Kreutzer correlations very large deviations are observed for higher values of pressure drop.

3.5. The homogeneous model

For the homogeneous model, the selection of methods for calculating density and viscosity of a homogenous mixture is the most important thing. In this paper, density was calculated from Eq. (10). To select the correlation for viscosity, which would give the best agreement with the experimental data, viscosity was calculated from Eqs. (11-13). The friction coefficient was calculated using Eq. (14) and the frictional pressure drop was calculated using Eq.(6). The calculated values of the frictional pressure drop were then compared with the experimental values. Table 4 shows standard deviation values for the ethanol-nitrogen system in the glass microreactor.

Table 4. The values of standard deviation for the considered viscosity correlations in the homogenous model (MPa/m) for ethanol-nitrogen system in the glass microreactor

$\mu \rightarrow$	Owens (1961) Eq. (11)	Dukler et al. (1964) Eq. (12)	Beattie and Whalley (1982) Eq. (13)
Homogenous model	0.074	0.024	0.087

As can be seen, the best results are obtained using Dukler's (1964) correlation for μ . Similar results were obtained for other physico-chemical systems and microreactors. In the further considerations the frictional coefficient was therefore calculated using Dukler's correlation for viscosity. For density the Kawahara correlation (Eq. 10) is used as mentioned earlier. The values of standard deviations are shown in Table 5.

It is seen that the homogenous model Eqs. (6,14) gives similar results as the classical model with the standard correlation for f (Eqs. 5, 7). For density and viscosity the formulas of Kawahara (Eq. 10) and Dukler (Eq. 12) should be used.

Table 5. The values of standard deviation for the considered frictional coefficient correlations in the homogenous model (MPa/m)

system	Homogenous model Eqs. (6, 14)
Glass Ethanol-nitrogen	0.024
Glass Heptane-nitrogen	0.017
Glass Water-nitrogen	0.047
PDMS Ethanol-nitrogen	0.111
Average	0.050

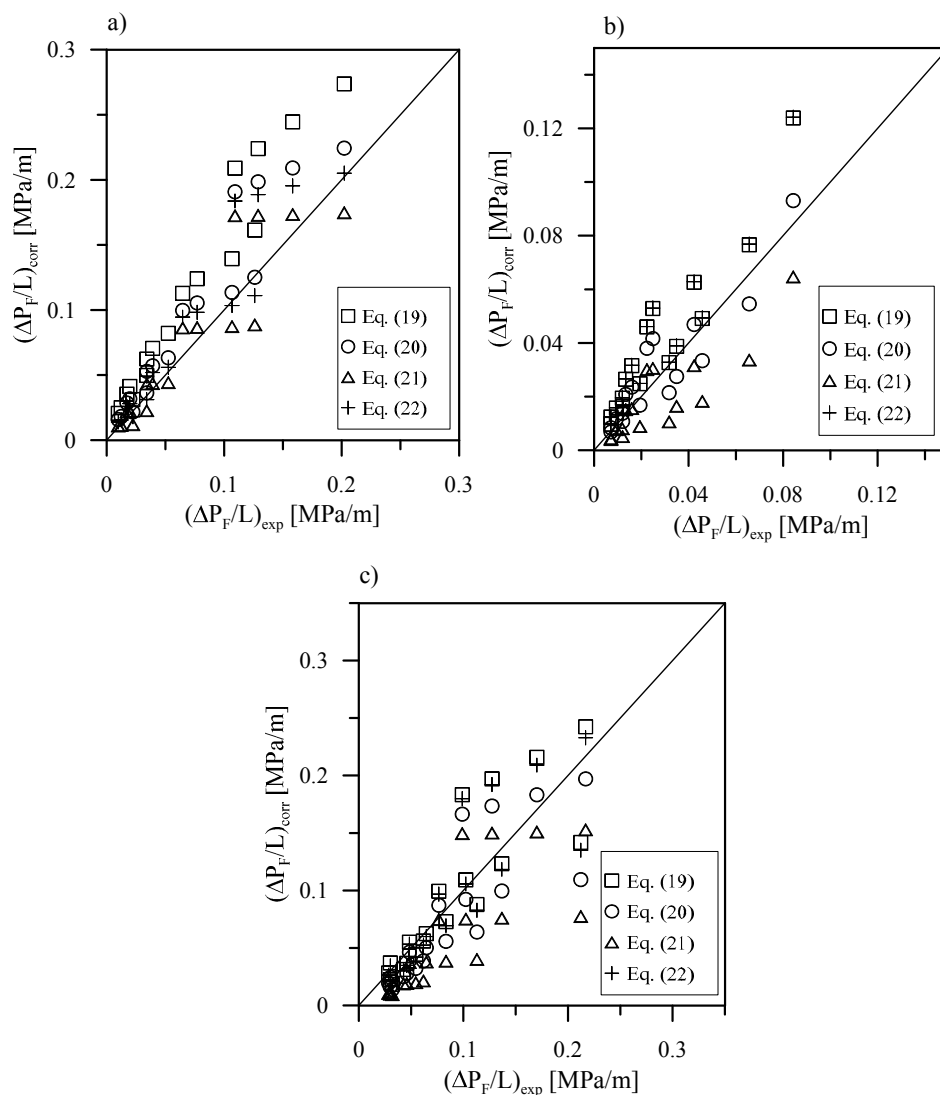


Fig. 12. Comparison of the experimental data with the existing correlations (Eqs. 19-22) for the frictional pressure drop in the glass microchannel: a) ethanol, b) heptane, c) water. Standard correlation (Eq. 7) was used for f

3.6. The Lockhart-Martinelli model

The experimental data have been compared with those calculated using the Lockhart-Martinelli model together with the existing correlations for the factor f (Eq. 7) and the factor C Eqs. (19-22). Fig. 12 shows the parity plots for the glass microchannel, Fig. 13 shows the parity plot for the PDMS microchannel (in all three PDMS microreactors the main microchannels were identical).

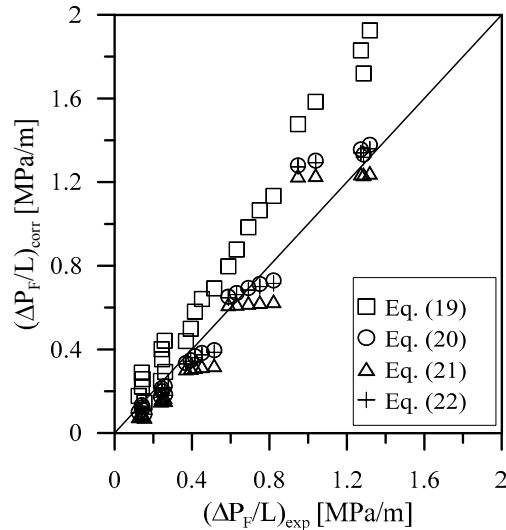


Fig. 13. Comparison of the experimental data with the existing correlations (Eqs. 19-22) for the frictional pressure drop in the PDMS microchannel for ethanol. Standard correlation (Eq. 7) was used for f

A comparison of our experimental data with those calculated from the Lockhart-Martinelli model using the existing correlations shows a generally reasonable agreement, except for the Chisholm (1967) correlation. This is understandable, as this correlation was developed for bigger channel diameters. Tab. 6 shows the values of the standard deviation for the data shown in Figs. 12-13 (Lockhart – Martinelli model). The results are somewhat better than those obtained for the classical model using standard (Eq. 7) correlation for the f coefficient, but the difference is not as large as it might have been expected (taking into account the assumptions of the classical model, rather far from reality for Taylor flow).

Table 6. The values of standard deviation for the different correlations for coefficient C in Lockhart-Martinelli model (MPa/m)

system ↓ \ C →	Chisholm (1967)	Mishima and Hibiki (1996)	Lee and Lee (2001)	Saisorn and Wongwises (2010)
Glass Ethanol	0.042	0.027	0.028	0.022
Glass Heptane	0.043	0.028	0.012	0.015
Glass Water	0.036	0.034	0.042	0.032
PDMS ethanol	0.280	0.098	0.101	0.098
Average	0.100	0.047	0.046	0.0432

On the basis of our experimental data we proposed our own correlations for the C factor:

$$C = 0.71 \cdot \lambda^{-0.233} \cdot Ca^{-0.024} \quad (24)$$

and for the friction factor:

$$f = \frac{14.015}{Re^{1.054}} \quad (25)$$

The parity plot of the results obtained for all the microchannels and all the systems using this correlation is shown in Fig. 14.

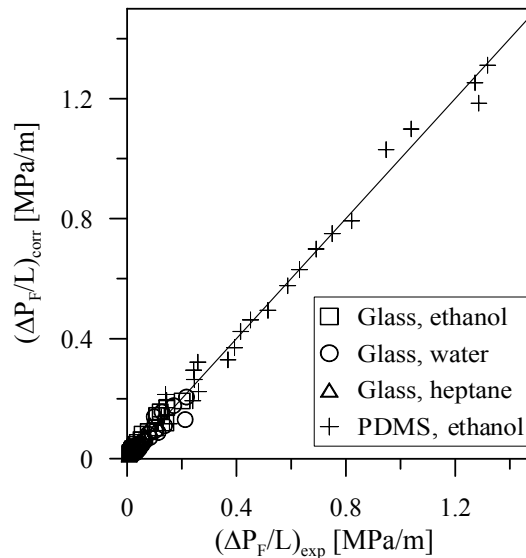


Fig. 14. Parity plot for the Lockhart-Martinelli model with own correlations for C and f (Eqs. 24-25)

The standard deviation for all the experimental points is 0.025 MPa/m, which shows a good fit of the proposed correlations to the experimental results.

4. CONCLUSIONS

The pressure drop in the Taylor gas-liquid flow in microchannels was measured. Four different microreactors and four different liquids were used. A 10 - 40% contribution of the inlet pressure drop to the total microreactor pressure drop was observed. Having eliminated the inlet effect, the pressure drop in the microchannel was determined. The experimental results were compared with the classical, homogenous and the Lockhart-Martinelli models. For each model a selection of correlations used was made.

For the classical model the best agreement with the experiments was obtained using the standard correlation (Eq. 7) for the friction coefficient f .

For the homogenous model; comparable results were obtained using the correlation (14) for f and the formulas of Kawahara (Eq. 10) and of Dukler (Eq. 12) for the density and viscosity of the mixture.

For the Lockhart-Martinelli model the best agreement was obtained using the standard (Eq. 7) correlation for the friction coefficient f , and the Saisorn and Wongwises correlation (Eq. 22) for the C factor.

Generally, the Lockhart-Martinelli model gave the best agreement with the experiment, but differences between the described models was not large, provided properly selected correlations for the model parameters were used. Our own correlations for the Lockhart-Martinelli model parameters were also proposed.

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SYMBOLS

C	factor in the Lockhart-Martinelli model, -
$Ca = \mu_L u_{TP} / \sigma$	capillary number, -
$Ca_H = \mu_{TP} u_{TP} / \sigma$	capillary number, -
d	hydraulic diameter, m
f	friction factor, -
g	gravity acceleration, $m \cdot s^{-2}$
G	total (gas and liquid) mass flux, $kg \cdot s^{-1} \cdot m^{-2}$
L	microchannel length, m
N	number of experiments, -
ΔP_Σ	overall measured pressure drop (inlet and total pressure drop), MPa
ΔP_A	pressure drop due to acceleration, MPa
ΔP_B	pressure drop over a moving bubble due to the change in shape between the bubble front and bubble tail, MPa
ΔP_F	frictional pressure drop, MPa
ΔP_G	pressure drop due to gravity, MPa
ΔP_{inlet}	inlet measured pressure drop, MPa
ΔP_T	total pressure drop in microchannel, MPa
$Re = ud\rho/\mu$	Reynolds number, -
$Re_H = u_{TP} d \rho_{TP} / \mu_{TP}$	Reynolds number, -
u	superficial velocity, $m \cdot s^{-1}$
V_b	volume of the bubbles in microchannel, m^3
V_s	volume of the liquid slugs in microchannel, m^3
x	gas mass fraction, -
X	Lockhart-Martinelli parameter, -

Greek symbols

α	channel slope angle, deg
$\beta = V_b / (V_s + V_b)^{-1}$	gas hold-up relative to phases volume, -
$\varepsilon_G = u_G u_{TP}^{-1}$	gas hold-up, -
$\lambda = \mu_L^2 \cdot \rho_L^{-1} \cdot \sigma^{-1} \cdot d_h^{-1}$	parameter, -
ϕ	two-phase multiplier in the Lockhart-Martinelli model, -
ρ	density, $kg \cdot m^{-3}$
μ	viscosity, Pa·s
σ	surface tension, $N \cdot m^{-1}$

Subscript

G	gas
H	homogenous model
L	liquid
TP	two phase

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