ELSEVIER

Chemical Engineering Journal 135S (2008) S185–S190

Chemical Engineering Journal

www.elsevier.com/locate/cej

Hydrodynamic regimes of gas–liquid flow in a microreactor channel

R. Pohorecki *, P. Sobieszuk, K. Kula, W. Moniuk, M. Zieliński, P. Cygański, P. Gawiński

Warsaw University of Technology, Faculty of Chemical and Process Engineering, Poland

Abstract

The hydrodynamic regimes of gas–liquid flow in a microreactor channel have been investigated, using water and ethanol (liquids with different contact angle and surface tension values) and nitrogen as an inert gas. The microreactor was Y-shaped. Experiments were carried out to determine the character of gas and liquid flow ("regime map"). Four main flow types were observed: bubble, slug, slug/annular and annular, the slug regime occupying most of the region investigated. In the second part of the work a criterion for the effectiveness of the gas–liquid interfacial area has been proposed. This criterion determines the mass transfer activity of the interfacial area in the slug regime. Methods of estimation of the parameters necessary to apply the criterion have been outlined.

Keywords: Microreactor; Interfacial area; Gas absorption; Chemical reaction

1. Influence of the interfacial parameters on the regime map

The aim of this part was to compare the flow regime maps for the microreactor gas–liquid flow, obtained using liquids with different contact angle and surface tension values. According to the literature suggestion [1], in a small diameter channel an influence of the above-mentioned parameters on the flow character should be observed. Barajas and Panton [1] have investigated the influence of the contact angle on the character of flow in a microchannel, however, they used a channel with rather large diameter (over 1 mm). They found significant differences between flow maps obtained for liquids with contact angle below and above 90°, but below 90° the influence of the actual value of the contact angle was only slight.

In this paper a channel with a few 100 μm diameter was investigated. Water and ethanol were used, with nitrogen as an inert gas. The flow regime was determined on the basis of films taken with a high-speed camera. The influence of the gas and liquid velocities were investigated for both liquids. The observed regimes have been classified as belonging to one of four kinds: bubble, slug, annular and slug/annular. The results have been compared with the "universal map" proposed by Vaillancourt et al. [2].

1.1. Experiments

The microreactor was designed for visual investigations of the hydrodynamic regime in two-phase gas–liquid system. The microreactor was Y-shaped with the following main channel dimensions:

Width	0.2 mm
Height	0.55 mm
Length	55 mm

The microreactor was made of PETg—polyethylene terephthalate modified by glycol. The experimental set-up is shown in Fig. 1. The gas nitrogen was supplied from a cylinder through a reducing valve and the needle valve. The gas flow has been split into two parts. One part led to the microreactor via flowmeter ("soap film meter"), the second one enabled monitoring gas pressure. The liquid flowed from the Mariotte's bottle by gravity. At the outlet from the microreactor the flow of liquid was measured by volumetric method.

Two liquids, water and ethanol, were used for the investigations. Those liquids have different values of surface tension and contact angle (Table 1).

The flow regime in the microreactor channel was recorded by CCD high-speed camera (PCI 8000S).

1.2. Results and discussion

In microreactor channel four main flow regimes were observed, as widely presented in the literature [3,4]: bubble, slug,

---

* Corresponding author at: Faculty of Chemical and Process Engineering, Warsaw University of Technology, Waryńskiego 1, PL 00-645 Warsaw, Poland. Tel.: +48 22 8258564; fax: +48 22 8258564.

E-mail address: pohorecki@ichip.pw.edu.pl (R. Pohorecki).

1385-8947/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.cej.2007.07.039

Nomenclature

b	stoichiometric coefficient
c	concentration (mol m^{-3})
$Ca = \mu u/\sigma$	capillary number
D	diffusivity (m^2 s^{-1})
E_i	enhancement factor
L	bubble length (m)
N	rate of absorption (mol m^{-2} s^{-1})
Q	absorbed quantity (mol m^{-2})
R	channel radius (m)
S	channel cross-section area (m^2)
t	time (s)
u	velocity (m s^{-1})
$\dot{V}$	volumetric flow rate (m^3 s^{-1})
x	linear coordinate (m)

Greek letters

β	solution of Eq. (17), parameter in Eqs. (13)–(16)
δ	depth of liquid layer (m)
λ	position of the reaction plane (m)
σ	surface tension (N m^{-1})
τ_C	contact time (s)
τ_R	minimal saturation/exhaustion time (s)

Subscripts

A	gaseous component
B	liquid component
g	gas
i	value at the interface
L	liquid
0	initial value

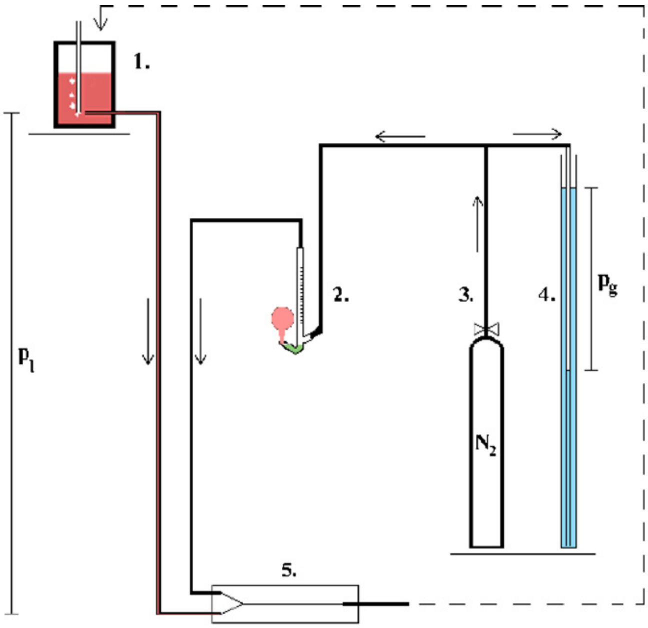

Fig. 1. The experimental set-up: (1) Mariotte's bottle; (2) gas flowmeter; (3) gas cylinder with the needle valve; (4) manometer "pipe in the pipe"; (5) microreactor.

Table 1
The properties of water and ethanol at 20 °C

Liquid	Contact angle (°)	Surface tension ($\times 10^{-3}$ N/m)
Water	69	72.9
Ethanol	0	23.5

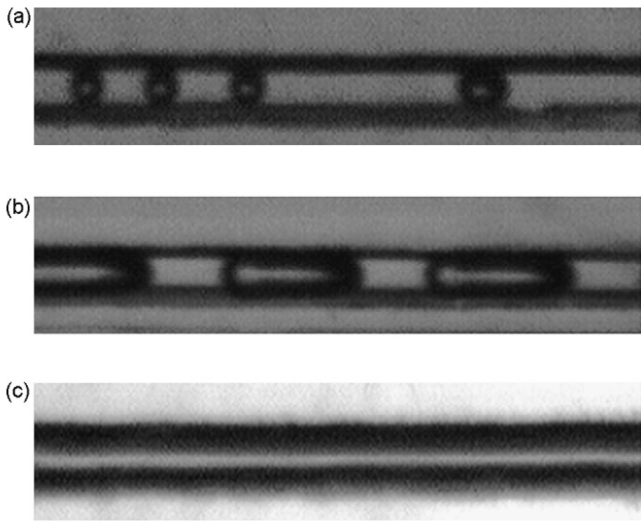

Fig. 2. Different flow characters for water–nitrogen set (a) bubble, (b) slug, (c) annular.

annular and slug/annular. Exemplary observed flow regimes for water–nitrogen system are shown in Fig. 2.

Observe different regime regions are shown in Figs. 3 and 4. The character of those maps is in qualitative agreement with the existing regime maps (lines) proposed by Vaillancourt et al. [2] and other authors [5–7] for channels with different diameters.

The influence of the contact angle and surface tension is evident, but not too large. For higher values of those parameters the line dividing the slug region and the annular region is displaced to the left towards lower values of apparent gas velocity. This effect was also observed by Barajas and Panton [1]. However, it should be pointed out that for contact angle values close and higher than 90° the flow becomes unstable and obtaining the regular slug regime becomes impossible.

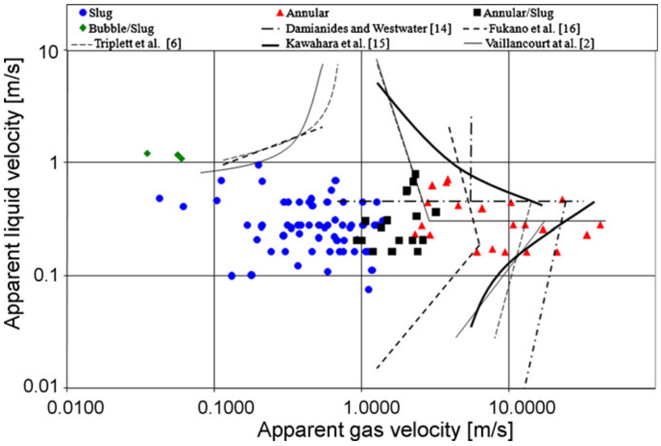

Fig. 3. The regime map for water–nitrogen system.

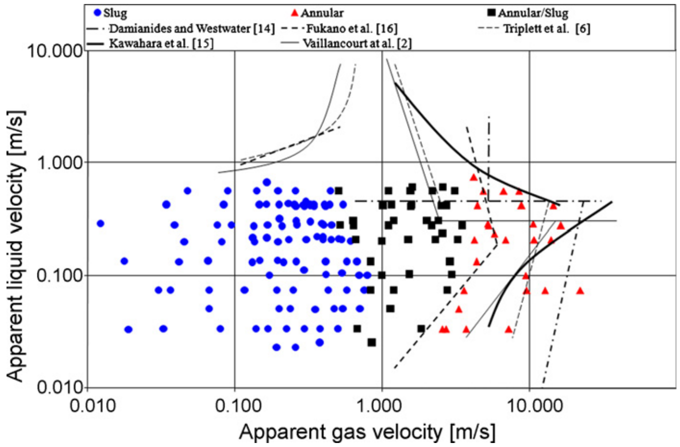

Fig. 4. The regime map for ethanol–nitrogen system.

2. Interfacial area in the slug regime [17]

As can be seen from the regime maps, the most common regime is the slug regime. In this regime slugs of one phase flow through the microchannel alternately with slugs of the other phase (gas bubbles in the case of a gas–liquid system). In what follows, the case of gas–liquid system shall be considered, although the conclusions apply also to the liquid–liquid system. If the liquid exhibits good wettability of the channel wall (i.e. low contact angle), then the wall is covered by a thin liquid layer, and the gas bubbles are sliding over this "lubricating" layer [8,9]. The interfacial gas–liquid area is then composed of two parts: the lateral part (i.e. that of the "lubricating" layer), and the perpendicular part (between the bubble and the adjacent liquid plug). As the length of the gas bubble is often many times greater than the channel diameter, the "lateral" part on the interfacial area may be many times greater than the "perpendicular" part (the lateral part contributes often more then 90% of the total interfacial area). The lubricating layer is usually very thin (its depth is of the order of 10^{-6} m [10]), and can therefore get saturated with the absorbed component or exhausted of the liquid-phase reactant. In such a case, the lateral part of the interfacial area will become inactive. As interfacial area is the main parameter governing the mass transfer/reaction rate (the mass transfer coefficients are usually less variable), it is of vital importance for the design of a microreactor to determine conditions of activity of this part of interface. In this paper an attempt shall be made to establish a criterion for the activity of the lateral part of the interfacial area.

2.1. Simplifying assumptions

Let us consider a gas bubble between two liquids plugs, as shown in Fig. 5. The bubble is moving with linear velocity u, its length is L, and the thickness of the lubricating layer is δ.

We shall assume that the liquid forming the lubricating layer is immobile or in laminar flow, and its contact time with the gas is

$$\tau_C = \frac{L}{u} \tag{1}$$

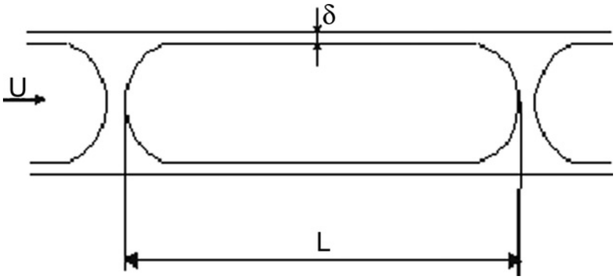

Fig. 5. Schematic view of gas–liquid flow in microchannel.

During this time the liquid absorbs a gaseous components which may (or may not) react with the liquid component. After that time the liquid is very quickly ("instantaneously") mixed with the content of the subsequent liquid plug (this, of course, is a serious simplification). We shall determine the conditions necessary to avoid saturation/exhaustion of the liquid forming the lubricating layer during the time span τ_C. To this end the exposition (contact) time, τ_C, should be much shorter than the saturation/exhaustion time τ_R ($\tau_C \ll \tau_R$)

2.2. Physical absorption

Physical absorption of a gas in an infinitely deep layer of quiescent liquid is described by the following equation:

$$D_A \frac{\partial^2 c_A}{\partial x^2} = \frac{\partial c_A}{\partial t} \tag{2}$$

with the boundary conditions

$$\begin{aligned} x &= 0 & t &> 0 & c_A &= c_{A_i} \\ x &> 0 & t &= 0 & c_A &= c_{A_0} \\ x &\to \infty & t &> 0 & c_A &= c_{A_0} \end{aligned} \tag{3}$$

A solution of this problem is known to be [11]

$$c_A - c_{A_0} = (c_{A_i} - c_{A_0}) \left(1 - \operatorname{erf} \frac{x}{2\sqrt{D_A t}}\right) \tag{4}$$

The rate of transfer, following from this solution, is

$$N_A = (c_{A_i} - c_{A_0}) \sqrt{\frac{D_A}{\pi t}} \tag{5}$$

and the amount absorbed by unit area of surface in time t is

$$Q = \int_0^t N_A \mathrm{d}t = 2(c_{A_i} - c_{A_0}) \sqrt{\frac{D_A t}{\pi}} \tag{6}$$

In the case of the liquid layer of a finite depth, the boundary conditions should be modified by replacing the condition for $x \to \infty$ by the conditions $\mathrm{d}c_A/\mathrm{d}x = 0$ at $x = \delta$. The solution takes then the form of a cosine series [12]. The initial absorption rate is, of course, identical as given by Eq. (5). For any finite period of time, the absorption rate into a layer of finite depth must be lower than that into an infinite layer. The shortest possible saturation time, τ_R, may therefore be estimated comparing the total absorbing capacity of the liquid layer, δc_{A_i} (where c_{A_i} is the saturation concentration of the gaseous component) with the amount absorbed into an infinitely deep layer

$$\delta c_{\mathrm{A}_i}=Q(\tau_{\mathrm{R}})=2c_{\mathrm{A}_i}\sqrt{\frac{D_{\mathrm{A}}\tau_{\mathrm{R}}}{\pi}} \tag{7}$$

(assuming that at $t=0$, $c_{\mathrm{A}_0}=0$). From Eq. (7) it follows that

$$\tau_{\mathrm{R}}=\frac{\pi}{4}\frac{\delta^2}{D_{\mathrm{A}}}\approx\frac{\delta^2}{D_{\mathrm{A}}} \tag{8}$$

2.3. *Absorption with an instantaneous chemical reaction*

For the case of absorption with an instantaneous irreversible chemical reaction

$$\mathrm{A}+b\mathrm{B}\rightarrow \text{products} \tag{9}$$

the following equations hold

$$D_{\mathrm{A}}\frac{\partial^2 c_{\mathrm{A}}}{\partial x^2}=\frac{\partial c_{\mathrm{A}}}{\partial t} \tag{10}$$

$$D_{\mathrm{B}}\frac{\partial^2 c_{\mathrm{B}}}{\partial x^2}=\frac{\partial c_{\mathrm{B}}}{\partial t} \tag{11}$$

with the boundary conditions (for an infinitely deep liquid layer)

$$\begin{array}{llll} x=0 & t>0, & c_{\mathrm{A}}=c_{\mathrm{A}_i} & \\ x=\lambda & t>0, & c_{\mathrm{A}}=0 & \\ x=\lambda & t>0, & c_{\mathrm{B}}=0 & \\ x\rightarrow\infty & t>0, & c_{\mathrm{B}}=c_{\mathrm{B}_0} & \\ x>0 & t=0, & c_{\mathrm{A}}=0, & c_{\mathrm{B}}=c_{\mathrm{B}_0} \\ x=\lambda & bN_{\mathrm{A}}=N_{\mathrm{B}} & & \end{array} \tag{12}$$

A solution to the above equation system has been given by Danckwerts [13] in the form

$$\frac{c_{\mathrm{A}}}{c_{\mathrm{A}_i}}=\frac{\operatorname{erfc}(x/2\sqrt{D_{\mathrm{A}}t})-\operatorname{erfc}(\beta/\sqrt{D_{\mathrm{A}}})}{\operatorname{erf}(\beta/\sqrt{D_{\mathrm{A}}})} \tag{13}$$

$$\frac{c_{\mathrm{B}}}{c_{\mathrm{B}_0}}=\frac{\operatorname{erf}(x/2\sqrt{D_{\mathrm{B}}t})-\operatorname{erf}(\beta/\sqrt{D_{\mathrm{B}}})}{\operatorname{erfc}(\beta/\sqrt{D_{\mathrm{B}}})} \tag{14}$$

where

$$N_{\mathrm{A}}=\frac{c_{\mathrm{A}_i}}{\operatorname{erf}(\beta\sqrt{D_{\mathrm{A}}})}\sqrt{\frac{D_{\mathrm{A}}}{\pi t}} \tag{15}$$

$$Q_{\mathrm{A}}=\frac{2c_{\mathrm{A}_i}}{\operatorname{erf}(\beta/\sqrt{D_{\mathrm{A}}})}\sqrt{\frac{D_{\mathrm{A}}t}{\pi}} \tag{16}$$

where β is given by

$$\mathrm{e}^{\beta^2/D_{\mathrm{B}}}\operatorname{erfc}\left(\frac{\beta}{\sqrt{D_{\mathrm{B}}}}\right)=\frac{c_{\mathrm{B}_0}}{bc_{\mathrm{A}_i}}\sqrt{\frac{D_{\mathrm{B}}}{D_{\mathrm{A}}}}\,\mathrm{e}^{\beta^2/D_{\mathrm{A}}}\operatorname{erf}\left(\frac{\beta}{\sqrt{D_{\mathrm{A}}}}\right) \tag{17}$$

Introducing the enhancement factor E_i

$$E_i=\frac{Q(\text{instantaneous reaction})}{Q(\text{physical})} \tag{18}$$

and noticing, that for an instantaneous reaction $E_i\gg 1$, one can write [11]

$$E_i\approx\sqrt{\frac{D_{\mathrm{A}}}{D_{\mathrm{B}}}}+\frac{c_{\mathrm{B}_0}}{bc_{\mathrm{A}_i}}\sqrt{\frac{D_{\mathrm{B}}}{D_{\mathrm{A}}}}\approx\frac{c_{\mathrm{B}_0}}{bc_{\mathrm{A}_i}}\sqrt{\frac{D_{\mathrm{B}}}{D_{\mathrm{A}}}} \tag{19}$$

For a liquid layer of a finite depth one should modify the boundary conditions by introducing an equation describing the exhaustion of the liquid component B

$$\int_{\lambda}^{\delta}c_{\mathrm{B}}\mathrm{d}x=c_{\mathrm{B}_0}\delta-bQ_{\mathrm{A}} \tag{20}$$

$$x=\delta,\qquad \frac{\mathrm{d}c_{\mathrm{B}}}{\mathrm{d}x}=0 \tag{21}$$

There is no simple solution to such a problem. However, by similar reasoning as for the case of physical absorption, one can estimate the shortest possible time necessary for the exhaustion of the liquid reactant comparing the total amount if this reactant in the liquid with the maximum consumption of this reactant

$$\delta c_{\mathrm{B}_0}=bQ_{\mathrm{A}}=2bE_ic_{\mathrm{A}_i}\sqrt{\frac{D_{\mathrm{A}}t}{\pi}} \tag{22}$$

$$\delta c_{\mathrm{B}_0}=2bc_{\mathrm{A}_i}\frac{c_{\mathrm{B}_0}}{bc_{\mathrm{A}_i}}\sqrt{\frac{D_{\mathrm{B}}}{D_{\mathrm{A}}}}\sqrt{\frac{D_{\mathrm{A}}t}{\pi}} \tag{23}$$

$$\tau_{\mathrm{R\,min}}=t=\frac{\pi\delta^2}{4D_{\mathrm{B}}}\approx\frac{\delta^2}{D_{\mathrm{B}}} \tag{24}$$

It is seen that in both cases, the physical absorption and the instantaneous reaction, the criterion of total surface area activity takes analogous form

$$\tau_{\mathrm{C}}=\frac{L}{u}\ll\tau_{\mathrm{R}}\approx\frac{\delta^2}{D} \tag{25}$$

where D equals D_{A} for the physical absorption case, or D_{B} for the instantaneous reaction case.

2.4. *Absorption with slow, solid catalysed reaction*

In the case of a reaction catalysed by a solid catalyst deposited on the surface of the channel walls, one is interested in an effect opposite to the described above: one would strive to maintain constant saturation of the liquid layer adjacent to the wall with the gaseous components. Such a situation may occur, for example, in solid catalysed hydrogenation reactions. It is obvious, that to assure constant saturation of the liquid layer, the reverse criterion should hold, namely

$$\frac{L}{u}\gg\frac{\delta^2}{D} \tag{26}$$

or

$$\frac{LD}{u\delta^2}\gg 1 \tag{27}$$

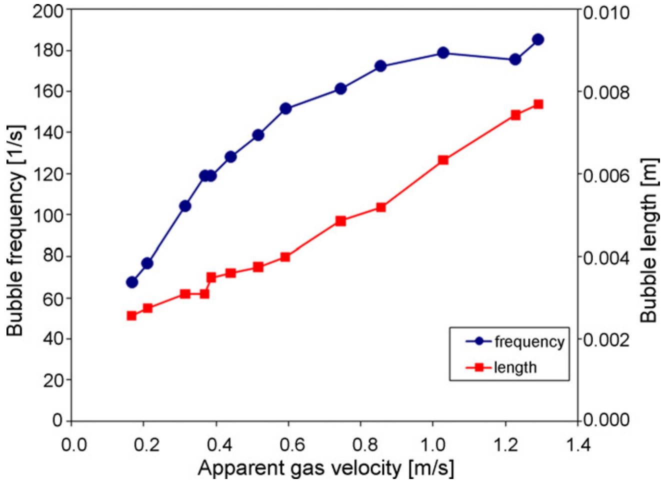

Fig. 6. Water–nitrogen system, bubble length and frequency, apparent liquid velocity: 0.28 m/s.

2.5. *Estimation of parameters*

To be able to apply the above criterion, one has to estimate the values of the parameters involved.

Diffusivities can be estimated by standard methods.

The thickness of the lubricating layer, δ, can by estimated using an empirical correlation of the form [10]

$$\frac{\delta}{R} = 1.0\, Ca^{1/2} \tag{28}$$

or from the Bretherton theory [10]

$$\frac{\delta}{R} = 0.643\, Ca^{2/3} \tag{29}$$

where R is channel radius, and Ca is the capillary number

$$Ca = \frac{\mu u}{\sigma} \tag{30}$$

The bubble velocity, u, can be estimated as

$$u \cong \frac{\dot{V}_{\rm g} + \dot{V}_{\rm L}}{S} = u_{\rm g} + u_{\rm L} \tag{31}$$

This is a simplification, since if does not differentiate between“dynamic” and “static” liquid holdup (by “dynamic” we mean the liquid contained in slugs, by “static” that contained in the lubricating layer and, in the case of channels with rectangular cross-section, contained in the corner menisci). For estimation purposes this, however, is usually used in the literature.

The bubble length, L, cannot easily be determined, as there is no sufficient amount of experimental data. We measured bubble length and bubble frequency for a range of $u_{\rm L}$ and $u_{\rm g}$ values, using two liquids (water and ethanol), and the experimental set-up described above. The results are given in Figs. 6–9. Further investigations are under way, to provide grounds for more thorough discussion.

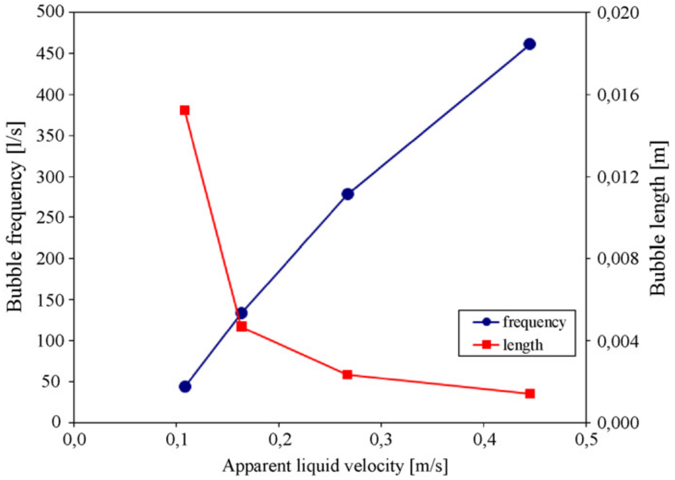

Fig. 7. Water–nitrogen system, bubble length and frequency, apparent gas velocity: 0.59 m/s.

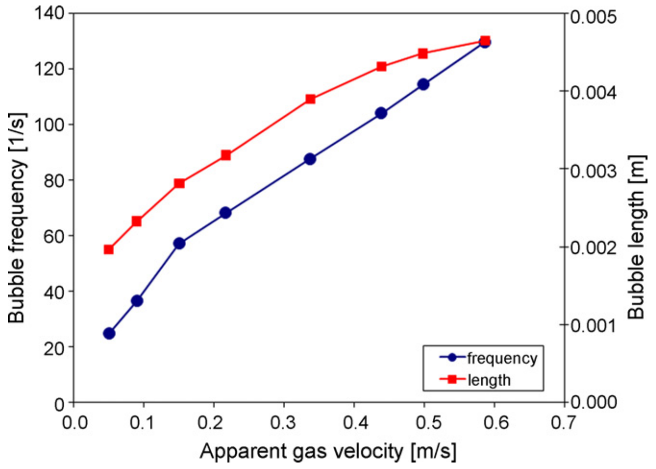

Fig. 8. Ethanol–nitrogen system, bubble length and frequency, apparent liquid velocity: 0.22 m/s.

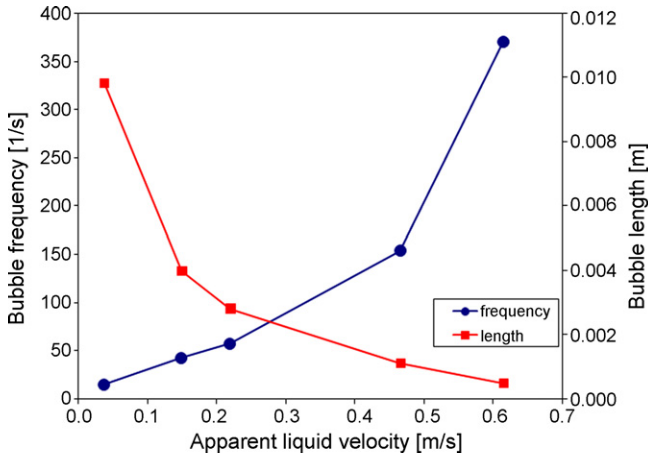

Fig. 9. Ethanol–nitrogen system, bubble length and frequency, apparent gas velocity: 0.16 m/s.

2.6. Discussion

Let us examine the typical microchannel flow to see whether the above-developed criteria may or may not be fulfilled. The order-of-magnitude estimations of relevant parameters are as follows

$$\delta \sim 10^{-6}\,\mathrm{m}$$
$$D_{\mathrm{B}} \sim 10^{-9}\,\mathrm{m^2/s}$$
$$\tau_{R\,\min} \sim 10^{-3}\,\mathrm{s}$$
$$L \sim 10^{-3}\,\mathrm{m}$$
$$u \sim 1\,\mathrm{m/s}$$
$$\frac{L}{u} \sim 10^{-3}\,\mathrm{s}$$

It can be seen, that both τ_{C} and τ_{R} values are of the order 10^{-3} s. It is therefore of vital importance to be able to determine not only gas (or liquid) holdup and interfacial area, but also size and frequency of gas bubbles flowing through a microchannel. It may be noticed, that the proposed criteria may also be applied to the annular regime case. For the annular regime the criterion should be determined using the total length of the channel and the thickness of the annular layer of liquid flowing along the channel walls.

3. Conclusion

The hydrodynamic regimes of gas–liquid flow in a microreactor channel have been investigated, using water and ethanol (liquids with different contact angle and surface tension values) and nitrogen as an inert gas. The influence of the contact angle and surface tension is evident, but not too large. For higher values of those parameters the line dividing the slug region and the annular region is displaced to the left towards lower values of apparent gas velocity. A criterion for effectiveness of the interfacial area in gas–liquid microreactors is proposed. The criterion takes the form

$$\frac{L}{u} \ll \frac{\delta^2}{D} \tag{32}$$

where D is equal D_{A} in the case of physical absorption, or D_{B} in the case of absorption accompanied by an instantaneous reaction. If this criterion is not fulfilled, one can expect severe decrease in the absorption/reaction rate. Methods of estimation of the parameters necessary to apply the criterion have been outlined.

Acknowledgement

This work has been supported by 6th Framework EU under Grant *IMPULSE* No. NMP2-CT-2005-011816.

References

[1] A.M. Barajas, R.L. Panton, The effect of contact angle on two phase flow in capillary tubes, Int. J. Multiphase Flow 19 (1993) 337–346.

[2] M.P. Vaillancourt, I.G. Hassan, K.K. Pehlivan, Two-phase flow transitions in microchannels. Proceedings of the 5th International Conference on Multiphase Flow, Yokohama, Japan 30.05-04.06.2004, Paper no. 181, 2004.

[3] A. Serizawa, Z. Feng, Z. Kawara, Two-phase flow in microchannels, Exp. Thermal Fluid Sci. 26 (2002) 703–714.

[4] W.L. Chen, M.C. Twu, C. Pan, Gas–liquid two-phase flow in microchannels, Int. J. Multiphase Flow 28 (2002) 1235–1247.

[5] P. Tortopidis, V. Bontozoglou, Mass transfer in gas–liquid flow in small-diameter tubes, Chem. Eng. Sci. 52 (1997) 2231–2237.

[6] K.A. Triplett, S.M. Ghiaasiaan, S.I. Abdel-Khalik, D.L. Sadowski, Gas–liquid two-phase flow in microchannels Part I: two-phase flow patterns, Int. J. Multiphase Flow 25 (1999) 194–377.

[7] M.K. Akbar, D.A. Plummer, S.M. Ghiaasiaan, On gas–liquid two-phase flow regimes in microchannels, Int. J. Multiphase Flow 29 (2003) 855–865.

[8] T. Cubaud, Ch.M. Ho, Transport of bubbles in square microchannels, Phys. Fluids 16 (2004) 4575–4585.

[9] W. Ehrfeld, V. Hessel, F. Löwe, Microreactors. New Technology for Modern Chemistry, Wiley–VCH, Weinheim, 2000, pp. 229–243.

[10] L.W. Schwartz, H.M. Princen, A.D. Kiss, On the motion of bubbles in capillary tubes, J. Fluid Mech. 172 (1986) 259–275.

[11] P.V. Danckwerts, Gas–Liquid Reactions, Mc Graw-Hill, New York, 1970, pp. 30–41.

[12] H.S. Carslaw, J.C. Jaeger, Conduction of Heat in Solids, Oxford University Press, Oxford, 1959, pp. 99–102.

[13] P.V. Danckwerts, Unsteady-state diffusion or heat conduction with moving boundary, Trans. Faraday Soc. 46 (1950) 701–712.

[14] C.A. Damianides, J.W. Westwater, Two-phase patterns in a compact heat exchanger and in small tubes, in: Proceedinds of the 2nd UK National Conference on Heat Transfer, 1988, pp. 1257–1268.

[15] A. Kawahara, P.M.Y. Chung, M. Kawaji, Investigation of two-phase flow patterns, void fraction and pressure drop in a microchannel, Int. J. Multiphase Flow 28 (2002) 1411–1435.

[16] T. Fukano, A. Kariyasaki, M. Kagawa, Flow patterns and pressure drop in isothermal gas–liquid coccurent flow in a horizontal capillary tube, in: ANS Proceedings of the National Heat Transfer Conference: Technical Session, 1989, pp. 153–161.

[17] R. Pohorecki, Effectiveness of interfacial area for mass transfer in two-phase flow in microreactors, Chem. Eng. Sci. 62 (2007) 6495–6498.