Instabilities of Immiscible Liquid-Liquid Two-Phase Laminar Flow in a Micro Channel

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Abstract

The present study investigated instabilities of immiscible liquid-liquid two-phase laminar flow in micro channel systems. Four representative flow patterns, i.e. stratified flow, stratified wavy flow, stratified flow with inclined interface and annular flow, were observed due to the difference of channel dimension of the organic-phase flow and physical properties of liquids.

Keywords: immiscible liquid-liquid flow, two-phase flow, micro channel, instabilities, flow visualization

1. Introduction

A micro channel chip is one of the most powerful tools for process intensification technology because of its favourable characteristics such as reduction of reagents, high throughput, energy and material saving and so on. The main advantages of micro channel chips due to decrease physical size are to intensify mass and heat transfer.

The immiscible liquid-liquid multi phase flow such as aqueous-organic phase flow in a micro channel system can be used for solvent extraction, interfacial chemical reaction, and drop formation. Nishisako *et al.* (2004) used a T-shaped micro channel to form monodispersed droplets. They reported that the droplet size could be varied flexibly in the range of diameters 30–120 by changing the flow conditions. Kawakatsu *et al.* (1999) proposed a novel method for producing monodispersed emulsion droplets from a microfabricated channel (MC) array. They call this technique MC emulsification. Sugiura *et al.* (2001) analyzed a drop-formation mechanism from a MC. They revealed that the interfacial tension between dispersed and continuous phases played an important role for droplet formation and stabilization. Sotowa *et al.* (2004) observed behaviours of a two-liquid flow in an X-shaped channel system. They conducted of styrene and methylmethacrylate co-polymerization in this channel system. Muto *et al.* (2004) used an X-shaped channel as a micro extractor to extract copper ions.

In order to intensify processes using liquid-liquid micro multiphase laminar flow, it is necessary to establish a precise control method. The present study investigated instabilities of immiscible liquid-liquid two-phase laminar flow in a micro channel.

2. Experimental

Figure 1 shows a schematic of the experimental setup. Syringes (Hamilton Company) and multiple syringe pumps (As One Corporation) were used for supplying the work fluids. The micro channel was linked to syringes through Teflon tubes (2 mm inner diameter). The effluent flow was discharged through the same Teflon tube. A digital micro scope (Keyence Corporation) was used to observe the behaviours of the immiscible two-phase flow. Visual data obtained the micro scope were taken in a personal computer. Figure 2 shows schematics of T-shaped channels. The two T-shaped channels of glass were fabricated by micro precision machining. The aqueous-phase-flow channel is 320 μm in width (d) and 120 μm in depth in both channel systems. One of the channel systems has the organic-phase-flow channel with 100 μm in width (d_1) and 120 μm in depth (Channel-1), while the other has the one with 320 μm in width and 120 μm in depth (Channel-2). The organic phase was fed into the channel through inlet A and the aqueous phase was fed into the channel through inlet B. Table 1 shows properties of the work fluids. An aqueous solution of glycerol or distilled water with or without surfactant was used as aqueous phase, while styrene monomer was used as organic phase.

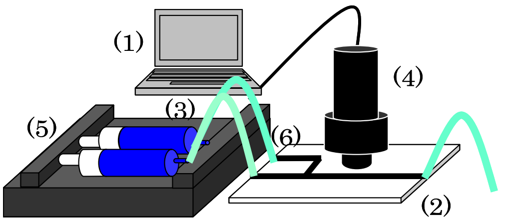

Fig. 1 Experimental setup; (1) personal computer, (2) micro channel, (3) syringe, (4) micro scope, (5) syringe pump and (6) silicone tube

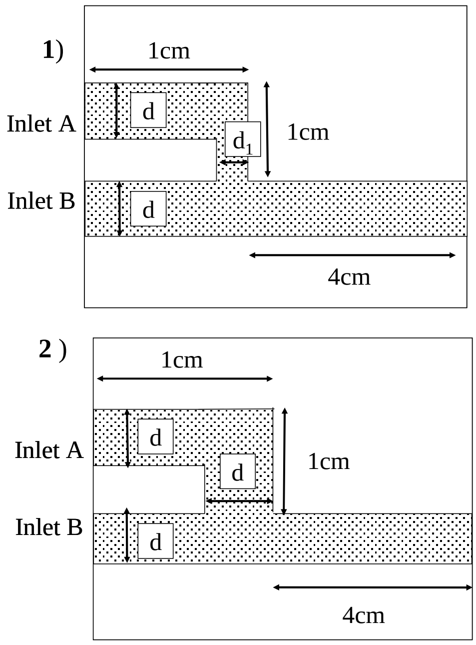

Fig.2 A diagram of details T-shaped channels

Both the interfacial tension between two fluids

Table.1 Properties of work fluid

Fluids	Density [kg/m^3]	Viscosity[Pa・s]	Interfacial tension[N/m]	Surface tension [N/m]
80vol% aquous solution of glycerol	1217.42	0.063461	0.0201	0.0623
70vol% aquous solution of glycerol	1193.18	0.027612	0.0214	0.0642
3wt% aqueous solution of SDS	1000.43	0.000990	0.0049	0.0371
styrene	911.00	0.000687	―	0.0310
distilled water	1000.00	0.001000	0.0339	0.0694

and the surface tension were measured by a Dunouy surface and interfacial tensiometer. The viscosity and the density were measured by a viscometer and a pyknometer, respectively. Owing to the difficulty of simultaneous supply of two fluids into the channel, the micro channel was initially filled with the aqueous phase Then both the fluids were fed into the channel. The flow rates of the aqueous phase (J_w) and the organic phase (J_s) are changed from 0.6m L/h to 10m L/h.

Numerical simulations were also conducted to investigate the velocity fields. A commercial CFD code RFLOW (RFLOW Co. Ltd) was used. The number of computational mesh is 8400. The geometry and boundary conditions for simulation were the same as those for the experiment. The velocity field was obtained by solving the three dimentional Navier-Stokes equations and the mass conservation for incompressible fluid based on the finite volume method.

3. Results and discussion

3.1. Flow patterns

Figure 3 shows four representative flow patterns, i.e. stratified flow, stratified wavy flow, stratified flow with inclined interface and annular flow, observed due to the difference of channel dimension of the organic-phase flow and physical properties of liquids. The stratified flows including the stratified wavy flow and the stratified flow with inclined interface were observed in the Channel-1. Stationary waves were superimposed on the interface when using pure distilled water, as shown in Figure 3 b). On the other hand, the straight interface was formed when adding surfactant (SDS) to distilled water (Figure 3 a)). The concentration of SDS was 3 wt% which was beyond the critical micelle concentration. This indicates that the formation of the non-wavy interface attributes to the decrease of the surface tension between two phases by adding an appropriate surfactant. In both the cases, the stratified flows formed

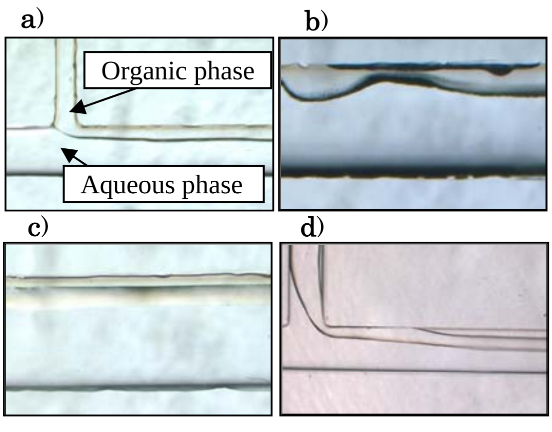

Fig.3 Flow patterns; a) stratified flow b) stratified wavy flow c) stratified flow with inclined interface and d) annular flow

perpendicular interface between two phases. When the aqueous phase of glycerol solution had relatively high viscosity (70-80 vol% of glycerol), not only the stratified flow but also a stratified flow with inclined interface was observed in the Channel-1 (Figure 3 c)). An annular flow was observed in the Channel-2 under a certain condition. The aqueous phase flowed as a film around the channel wall and a core of the organic phase formed in the middle of channel. In this flow pattern, the aqueous phase needs to be relatively high viscosity (70-80 vol% of glycerol). These results indicate that the channel dimensions, viscosity and surface tension affect flow instabilities.

3.2. Slip ratio and void fraction

In the present work, the void fraction α is defined as follows:

$$\alpha = \frac{A_s}{A_s + A_w} \tag{1}$$

where A_s and A_w are the cross-sectional area of organic and aqueous phases respectively. The slip ratio S is expressed by the following equation:

$$S = \frac{u_s}{u_w} \tag{2}$$

where u_s and u_w are the average velocity of organic and aqueous flows respectively. Then the void fraction can be estimated by the slip ratio S and the ratio of volumetric flow rates of organic phase, J_s, to the total volumetric flow rate, $J_s + J_w$.

$$\alpha = \left[1 + \left(\frac{J_w + J_s}{J_s} - 1\right)S\right]^{-1} \tag{3}$$

Figure 4 shows the void fraction against the ratio of volumetric flow rates of organic phase to the total volumetric flow rate. The solid lines in Figure 4 were numerically obtained by Eq. (3) . Correlation between α and $J_s/(J_s + J_w)$ depends on the viscosity difference between two fluids. When the lowest viscosity fluid in aqueous phase (water with 3 wt% SDS) was used, the viscosity difference between aqueous and organic phases was very small. In this case, α and $J_s/(J_s + J_w)$ could be correlated with the lower slip ratio ranging from 0.5 to 2.0. On the contrary, when the relatively higher viscosity fluid in aqueous phase (aqueous solution of glycerol) was used, they could be correlated with the higher slip ratio up to 20. Forthermore, in the case of stratified flow, α and $J_s/(J_s + J_w)$ could not be correlated by an unique slip ratio. The slip ratio became large with increasing volumetric flow rate of organic phase. On the other hand, in the case of

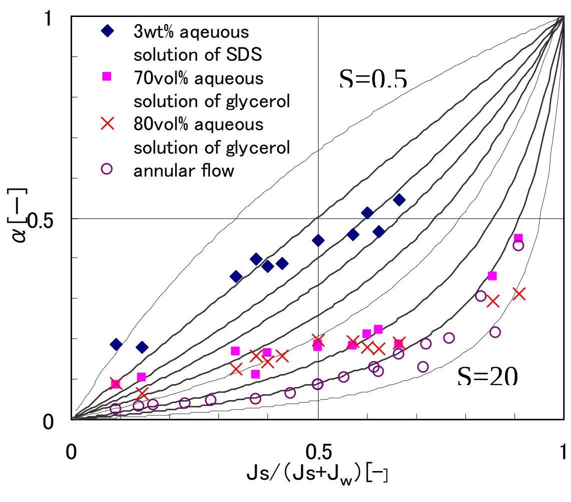

Fig.4 α against $J_s/(J_s+J_w)$

annular flow, α and $J_s/(J_s + J_w)$ could be correlated well by Eq. (3) with S = 10. This may be due to the fact that the oroganic phase exposes only the water phase and that there is no friction between the wall of channel and the organic flow. These results indicate that the wall effect of channel is not negligible in the case of stratified flow.

3.3. Characteristics of stratified flow

In order to characterize the stratified flows, the Lockhart-Martinelli method (Lockhart and Martinelli, 1949) was used. In the stratified flows, it can be assumed that the two-phase pressure gradient, the aqueous-phase pressure gradient and the organic-phase pressure gradient are equal to each other. In laminar flow, the Martinelli's parameter X can be described as the following equation:

$$X^2 = \frac{\left(\mathrm{d}P_{\mathrm{w}0} \middle/ \mathrm{d}z \right)}{\left(\mathrm{d}P_{\mathrm{s}0} \middle/ \mathrm{d}z \right)} = \frac{\mu_{\mathrm{w}}}{\mu_{\mathrm{s}}} \cdot \frac{J_{\mathrm{w}}}{J_{\mathrm{s}}} \tag{4}$$

where μ_s and μ_w are the viscosity of aqueous and organic phases respectively. Figure 5 shows the void fraction against Martinelli's parameter. The solid line was obtained by the Chisholm's approximation (Eq. (5)) (Chisolm, 1967) while the break line was obtained by the Wang's approximation (Eq. (6)) (Wang *et al.* 1990).

$$\alpha = 1 - \left(1 + \frac{5}{X} + \frac{1}{X^2} \right)^{-1/2} \tag{5}$$

$$\alpha = 1 - \left(1 + \frac{2.6}{X} + \frac{0.1}{X^2} \right)^{-1/2} \tag{6}$$

Figure 5 shows that the most of the experimental data exist in between these two approximations. When the viscosity difference between the aqueous and the organic phases is very small, the void fraction can be expressed by the Wang's approximation. On the other hand, when the viscosity difference between the aqueous and the organic phases is large, the data of void fraction tend to approach the Chisholm's approximation. The Chisholm's approximation is mainly applicable for gas-liquid two phase flow. It can be considered that characteristics of liquid-liquid two phase flow with larger viscosity difference are similar to those of gas-liquid two phase flow.

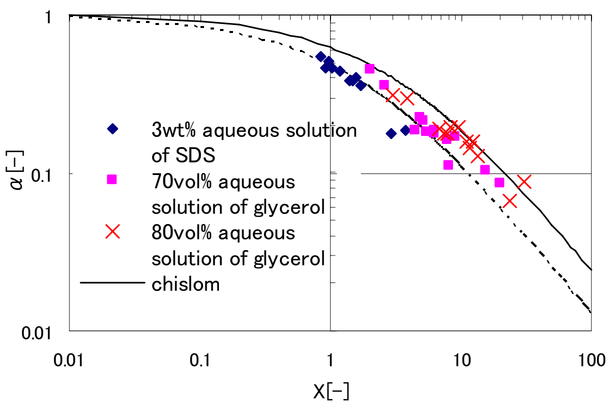

Fig.5 α of stratified flow against Martinelli's parameter X

3.4. Characteristics of annular flow

As described in the previous section 3.1, an annular flow of the aqueous solution was observed in the Channel-2 under a certain condition. The aqueous phase flowed as a film around the channel wall (wall flow) and a core of the organic phase formed in the middle of channel (core flow). In this flow pattern, the aqueous phase needs to be relatively high viscosity. In the annular flow, the void fraction can be obtained by the following equation (Fujii *et al*,. 1993)

$$\alpha = \frac{2}{\sqrt{\left(k\frac{J_w}{J_s}\right)^2 + X^2\frac{4}{B}} + k\frac{J_w}{J_s} + 2} \tag{7}$$

where k is a correction factor for the ratio of the volumetric flow rate of the aqueous phase to that of the organic phase, and B is a parameter for the roughness between two phases. The present work adopted $k = 1$. Figure 6 shows the void fraction α against Martinelli's parameter X. The void fraction obtained by the experiments shows quantitatively good agreement with Eq. (7) with $B = 1$. This result indicates that the interface between organic phase and aqueous phase was smooth without surface wave.

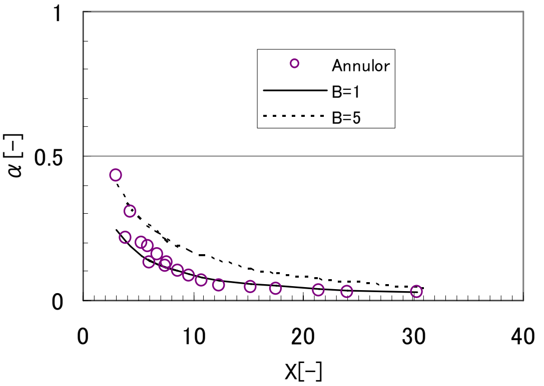

Fig.6 α of annular flow against Martinelli's parameter X

In the present work, no annular flow pattern could be observed and only the stratified flow patterns were observed in Channel-1. Figure 7 shows a photograph and a numerically-obtained velocity vector field of the annular flow in Channel-2. As can be seen in the circled region in Figure 7 a), the aqueous phase intrudes into the organic flow channel. Numerical result indicates that the flow stagnates in this region (Figure 7 b)). On the other hand, no such a stagnant region can be found in Channel-1. The present work set two-dimensional rectangular x-y coordinates and a reference coordinate r-axis (A-B) inclined 135° against the x-axis as shown in Figure 8. Here, the origin of the test line is point A. The velocity components (v_x, v_y) on the r-axis were numerically obtained. Figure 9 shows the ratio of v_y to v_x on the

a)

b)

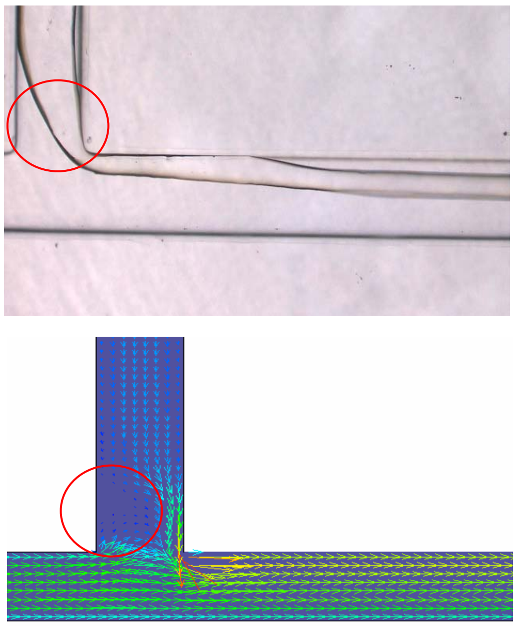

Fig.7 Annular flow pattern; a) experimental image and b) velocity field obtained by numerical simulation

r-axis. Figure 9 clearly shows that v_y/v_x is relative larger in Channel-2 than in Channel-1. As compared with v_y, v_x in the confluence region in Channel-2 is very weak. Then the relatively larger velocity component v_y detaches the organic phase from the channel wall. It has, therefore, been found that the flow in the confluent region affect the flow patterns.

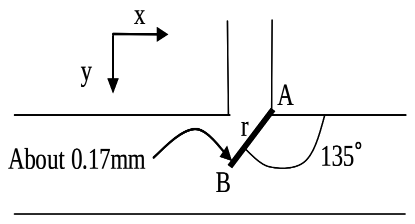

Fig.8 Coordinate system for numerical analysis of the annular flow

4. Conclusions

The present study investigated instabilities of immiscible liquid-liquid two-phase laminar flow in micro channel systems. The following conclusions can be deduced:

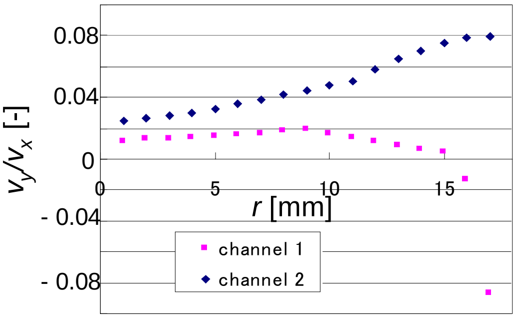

Fig. 9 V_y/V_x against *r*

1) Four representative flow patterns, i.e. stratified flow, stratified wavy flow, stratified flow with inclined interface and annular flow, were observed due to the difference of channel dimension of the organic-phase flow and physical properties of liquids.
2) The stratified flows were characterized by the Lockhart-Martinelli method, and the void fraction can be expressed by the Chisholm's approximation or the Wang's approximation.
3) The void fraction obtained by the annular flow shows quantitatively good agreement with Eq. (7) with B = 1. This result indicates that the interface between organic phase and aqueous phase was smooth without surface wave.
4) The channel dimensions, viscosity and surface tension affect flow instabilities.

Acknowledgements

The Authors wish to thank Prof. Kenichiro Sotowa of Tokushima University for manufacturing the micro channels. This research was financially supported by a Grant-in-Aid for Scientific Research (A) (No. 17206079) from the Japan Society for the Promotion of Science (JSPS).

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