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THE PRESSURE DROP AND DYNAMIC CONTACT ANGLE OF MOTION OF TRIPLE-LINES IN HYDROPHOBIC MICROCHANNELS

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ABSTRACT

At two-phase flow in microchannels, slug flow regime is different for wettability of surface. A slug in a hydrophilic microchannel has liquid film. However, a slug in a hydrophobic microchannel has no liquid film instead, the slug has triple-lines and makes higher pressure drop due to the motion of the triple-line. In previous researches, pressure drop of triple-line is depended of dynamic contact angle, channel diameter and fluid property. And, dynamic contact angle is depended of static contact angle, superficial velocity and fluid property. In order to understand the pressure drop of motion of triple-lines, pressure drop of slug with triple-lines in case of various diameters (0.546, 0.763, 1.018, 1.555, 2.075 mm), various fluids (D.I.water, D.I.water-1, 5, 10% ethanol mixture) and various superficial velocity (j=0.01~0.4 m/s) was measured. Dynamic contact angle was calculated from relation of the pressure drop of slug with triple-lines. Comparing with previous dynamic contact angle correlations, previous correlation underestimated dynamic contact angle in the region of this study. ($10^{-4} \leq Ca \leq 10^{-3}$, $10^{-2} \leq We \leq 10^{-1}$, $68° \leq \theta_S \leq 110°$)

NOMENCLATURE

P	pressure [Pa]
j	superficial velocity [m/s]
D	conduit diameter [m]
Ca	capillary number [dimensionless]
L	length [m]
C	coefficient
k_B	Boltzman constant [J/K]
T	absolute temperature [K]
We	Weber number

Greek symbols

Δ	difference
θ	contact angle [radian]
σ	surface tension [N/m]
μ	viscosity [Pa.s]
f	friction factor [dimensionless]
ρ	density [kg/m3]
κ°	characteristic frequency [s-1]
λ	characteristic length [m]

Subscripts

TL	triple-line
LG	liquid-gas phase
L	liquid phase
G	gas phase
TP	two-phase
GS	gas slug
LS	liquid slug
SP	single phase
D	dynamic
A	advancing (dynamic)
R	receding (dynamic)
S	static
m	molecular

1. INTRODUCTION

Two-phase flow regime in microchannels is influenced on wettability of channel [1]. Especially, according to wettability of channel, slugs of different form are generated: in hydrophilic microchannels, slugs with the liquid film are generated, but hydrophobic microchannels, slugs with triple-lines are generated. In the hydrophilic case, area change of liquid flow at nose and tail of slugs generate dominant two-phase pressure drop. However, motion of triple-lines generate dominant two-phase pressure drop. Comparing the pressure drop of each case, slug with triple-line generate higher pressure drop than slug with liquid film [2]. Because, comparing liquid slugs smoothly move on the liquid film in hydrophilic microchannels, liquid slugs move by dragging triple-lines on the dry surface in hydrophobic microchannel.

In the past, many researchers interested dynamic contact angle rather than pressure drop of triple-lines [3] ~ [8]. And, many researchers investigated dynamic contact angle and reported that dynamic contact angle was affected on static contact angle and capillary number. However, previous researches were carried out in low superficial velocity region (j=0~0.04m/s) than slug flow regime region (j=0.05~0.43m/s), because the fluid of oil type was used by working fluid in the experiments.

Recently, P.Rapolu et al. developed the equation of pressure drop of triple-line that adopted dynamic contact angle on Young's equation [9]. In the equation, pressure drop of triple-line affected on channel diameter, dynamic contact angle and fluid property. Lee and Lee was calculated pressure drop of triple-line using experimentally measured pressure drop of slug with triple-line. And, they developed dynamic contact angle correlation that enabled to predict pressure drop of triple-line with P.Rapolu et al.'s equation [10]. But, parameter study about pressure drop of triple-lines was not conducted yet. Also, understanding of dynamic contact angle was lack to develop the correlation about pressure drop of triple-lines in the slug flow regime region.

In this present research, we conducted the experimental study of parameters that affect on pressure drop of triple-lines based on P.Rapolu et al.'s equation. And, to understand of dynamic contact angle that is dominant parameters that affect on pressure drop of triple-lines, the previous researches of dynamic contact angle was investigated and compared with calculated dynamic contact angle using measured pressure drop of slug with triple-lines in the slug flow regime region.

2. EXPERIMENTS

2.1 EXPERIMENTAL CONDITION

P.Rapolu et al. suggested the equation of pressure drop of triple-line (ΔP_{TL}). According to the equation, ΔP_{TL} was affected on channel diameter, dynamic contact angle (θ_D) and fluid property. Many theoretical and experimental researches of θ_D were reported. The well-known theoretical analysis of θ_D was hydrodynamic model and kinetic molecular model [11]. Hydrodynamic model focuses on the viscous force in the wedge adjacent to moving triple-lines. This force is generated by viscous bending between the moving triple-line and the no-slip region. Kinetic molecular model focuses on the friction at the moving triple-line. This friction is generated by attachment and detachment of molecules in the triple-line. Each model was different approaches to θ_D. But, they have common opinion that θ_D was affected on static contact angle (θ_S), superficial velocity and fluid property. The opinion was accepted on the experimental analysis. Finally, according to above, ΔP_{TL} was affected on diameter and superficial velocity and fluid property.

$$\Delta \mathrm{P}_{\mathrm{TL}} = -\frac{4\sigma}{\mathrm{D}}\left(\cos\theta_{\mathrm{A}} - \cos\theta_{\mathrm{R}}\right) \quad (1)$$

$$\theta_{\mathrm{A}}^3 - \theta_{\mathrm{S}}^3 = 9Ca\ln\left(\frac{\mathrm{L}}{\mathrm{L_m}}\right) \quad (2)$$

$$j = 2\kappa^{\circ}\lambda\sinh\left[\sigma\left(\cos\theta_{\mathrm{A}} - \cos\theta_{\mathrm{S}}\right)\lambda^2/2k_{\mathrm{B}}\mathrm{T}\right] \quad (3)$$

To understand the parameters that were diameter, fluid property and superficial velocity, pressure drop of slug with triple-lines was measured in various diameters (0.546, 0.763, 1.018, 1.555, 2.075 mm), fluids (D.I.water, D.I.water-1, 5, 10% ethanol) and superficial velocity (0.01~0.43m/s). Experimental range of superficial velocity was based on slug flow regime region of Lee and Lee experimental data.

2.2 EXPERIMENT PREPARATION

In our experiments, two-phase flow was composed of nitrogen and D.I.water-ethanol mixture. Density, viscosity, surface tension and static contact angle of D.I.water-ethanol mixture were respectively measured using the electric balance (Sartorius, 1600S), the Cannon Fenske kinematic viscometer (Cannon, 50), the surface tension meter (ITOH, 514-A) and sessile drop method. And, values of measured properties were compared with previous researches [12] ~ [14]. To analysis fluid property effect, we adopted characteristic velocity that was defined in Eq.3 by de Genne [15]. Each property is summarized in Table.1.

$$\mathrm{j}^* = \frac{\sigma}{\mu},\ \mathrm{Ca} = \frac{\mu \mathrm{j}}{\sigma} = \frac{\mathrm{j}}{\mathrm{j}^*} \quad (4)$$

Before conducting the experiments, test sections were cleaned by the following methods. Organic matters in the test sections were cleaned to orderly inject acetone, methanol and D.I.water. Subsequently, the wet in the test section were cleaned to blow using nitrogen gun and make vacuous along 24 hours in chamber.

2.3 EXPERIMENTAL SETUP

Figure.1 is the schematic diagram of our experimental loop. Test sections were conventional round mini-microchannels (HANEOL, HE-02, UPCHURCH, 1512L / 1514L / 1507L, SWAGELOK, Q02309). Test sections were made on hydrophobic material (PFA: Perfluoroalkoxy). The length of test section was 8 m to secure the fully developed region.

Table.1 D.I.water-ethanol

Properties	ethanol mole fraction			
	0%	1%	5%	10%
ρ [kg/m^3]	997.05	997.27	977.08	963.38
σ [N/m]	0.073	0.064	0.046	0.038
μ [mPa.s]	0.1046	0.1160	0.1651	0.2186
j* [m/s]	69.86	55.55	27.94	17.35
θ_S [°]	110	85	77	68

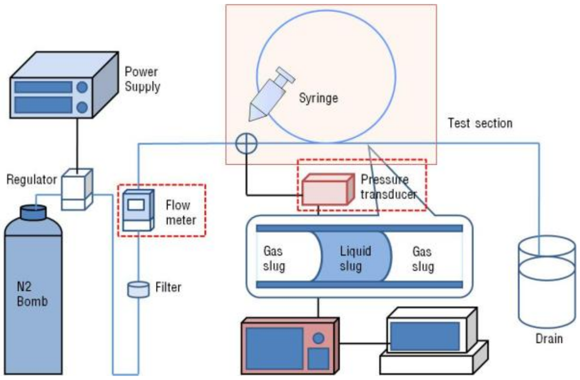

Figure.1 Schematic diagram

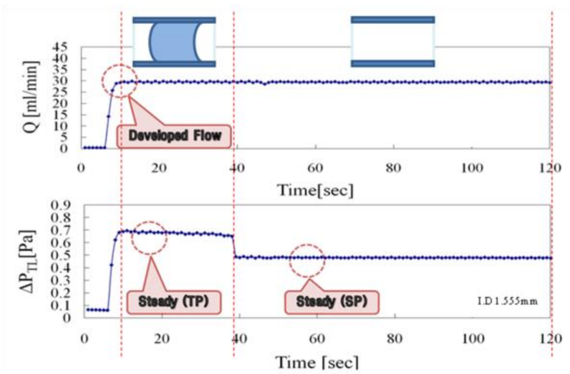

Figure.2 data reduction

Using the micro liter syringe, liquid slug was injected into the test sections. The length of test section was 8 m to secure the fully developed region. The length of liquid slug was larger than 5 times of channel diameters. Subsequently, the electronic regulators (SMC, ITV1010-211CS5 / ITV1030-211CS5) and the power supply (VUPOWER, AK-3003DD) controlled pressure of nitrogen bomb and made the steady two-phase flow. At upstream of the test section, the gas flow rate was measured using the gas mass flow meters (OMEGA, FMA-2615A / 2604A) and the pressure drop of two-phase flow and single phase flow were measured using the pressure transducers (SETRA, 209, SMC, PSE550). All measured data were gathered using personal computer and data acquisition system (GRAPHTEC, WS450) every second.

3. DATA REDUCTION

In experiments, pressure drop of two-phase (ΔP_{TP}), single phase (ΔP_{SP}) and gas flow rate were measured. In figure.2, when liquid slug passed the test section, ΔP_{TP} and gas flow rate were measured. Subsequently, when liquid slug went out the test section, ΔP_{SP} and gas flow rate were measured.

At slug with triple-lines, to conserve continuity equation, gas slug, liquid slug, triple-lines superficial velocity at the slug flow with triple-lines were equivalent because of gas slug with no liquid film. Also, geometrically, the length of the test section is equal to the sum of length of liquid slug and gas slug.

$$j_G = j_L = j_{TL} = j \tag{5}$$

$$L_T = L_L + L_G \tag{6}$$

ΔP_{TP} is composed of pressure drop of liquid slug (ΔP_{LS}), gas slug (ΔP_{GS}) and ΔP_{TP}. Therefore, ΔP_{TL} is equivalent that ΔP_{TP} subtracted ΔP_{LS} and ΔP_{GS} Friction factor was calculated using Eq.7

$$\Delta P_{TP} = \Delta P_{LS} + \Delta P_{GS} + \Delta P_{TL} \tag{7}$$

$$\Delta P_{TL} = \Delta P_{TP} - f_L \frac{L_L}{2D} \rho_L j^2 - f_G \frac{L_T - L_L}{2D} \rho_G j^2 \tag{8}$$

$$f_L \frac{\rho_L j D}{\mu_L} = f_G \frac{\rho_G j D}{\mu_G} = 64 \tag{9}$$

θ_A was calculated using pressure drop of triple-lines. First of all, we adopted Lee and Lee's assumption that difference with θ_S and advancing and receding dynamic contact angle (θ_A, θ_R) were similar at front and front of slug with triple-line. Finally, inserting Eq. 9 to Eq. 1, θ_A was calculated using Eq. 10.

$$\theta_A - \theta_S \approx \theta_R - \theta_S \tag{10}$$

$$\theta_A = \arccos\left[-\frac{C_1 C_2}{C_2^2 + C_3^2} \pm \sqrt{\left(\frac{C_1 C_2}{C_2^2 + C_3^2}\right)^2 - \left(\frac{C_1^2 - C_3^2}{C_2^2 + C_3^2}\right)} \right]$$

$$\left(C_1 = \frac{\Delta P_{TL} D}{\sigma}, C_2 = 4(1 - \cos\theta_S), C_3 = 4\sin\theta_S \right) \tag{11}$$

4. EXPERIMENTAL RESULTS

First of all, experiments were carried out to investigate whether $\Delta P_{GS,}$ ΔP_{LS} and ΔP_{TL} was independent of the length of liquid slug. In the test section (D=1.555mm), experiments were carried out in cases of various liquid lengths. (D.I.water, 0.5, 1.0, 2.0cm) ΔP_{TL} at different liquid lengths were equivalent within 4% in figure.3. ΔP_{TL} could be considered independent of the length of liquid slug.

ΔP_{TL} increased as channel diameter decreased (figure.4). This trend is agreement that ΔP_{TL} is inversely proportional to diameters in Eq.8.

ΔP_{TL} increased as superficial velocity increased (figure.5). According to Eq.1, ΔP_{TL} increases as θ_A increased, also θ_A increased as superficial velocity increased in Eq.2, 3. Eventually, ΔP_{TL} in the channel axis-direction increased as θ_A increased alike experimental results.

As ethanol mole fraction increased, density and surface tension of D.I.water-ethanol mixture decreased smoothly, but the viscosity of the mixture increased in the experimental range. Eventually, j* was decreased as ethanol mole fraction increased. Over the range of conditions used in this study, ΔP_{TL} decreased and the slopes of the relationship between ΔP_{TL} and j increased as j* decreased, however the slopes of the relationship between ΔP_{TL} and Ca similar in all j*. Through the results, we could know that ΔP_{TL} is well-correlated of Ca (figure.6)

θ_A of various fluids increased with similar slope as Ca increased, but θ_A differed among the various fluids because

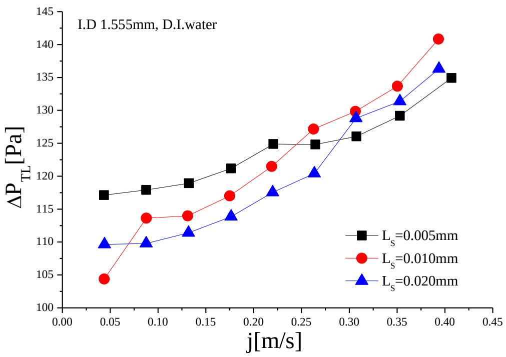

Figure.3 ΔP_{TL} of various lengths of liquid slug

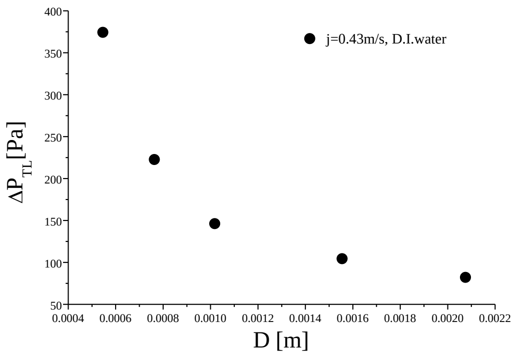

Figure.4 ΔP_{TL} of various channel diameters

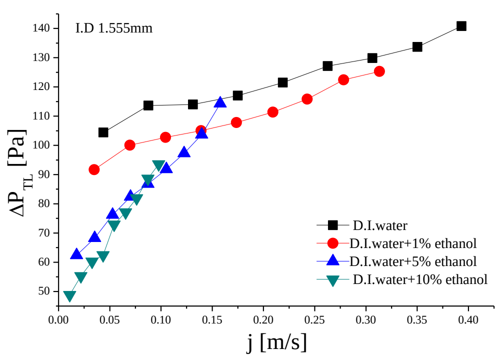

Figure.5 ΔP_{TL} of superficial velocity

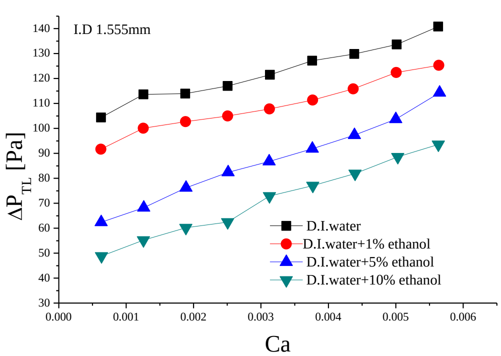

Figure.6 ΔP_{TL} of Ca

their θ_S were different. (figure.7) θ_A were independent of channel diameter. (figure.8)

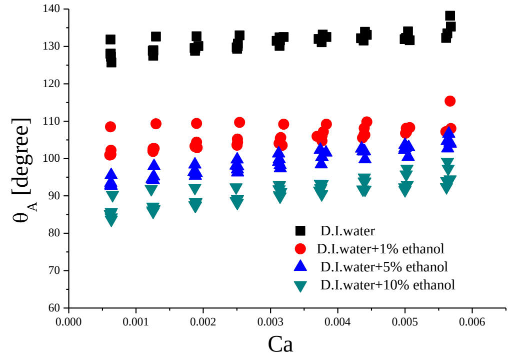

Figure.7 θ_A of Ca

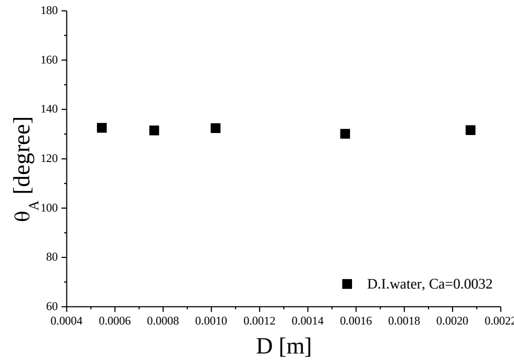

Figure.8 θ_A of various channel diameters

5. COMPARISON WITH PREVIOUS RESEARCHES

ΔP_{TL} of our research compared with Lee and Lee's data. Because, Lee and Lee were recently carried out the trailblazing research about ΔP_{TL} and θ_A. In a water-air-polyurethane system (θ_S =77°) and a water-air-teflon system (θ_S =110°), they measured ΔP_{TP} in the slug flow regime. Comparing with our data (D.I.water, D.I.water-5% ethanol), their data was agreed within 25%. (figure. 9)

θ_A of our research compared with previous θ_A correlations. Previous correlations have been based on the Jiang et al.'s universal correlation (H) (Eq. 11). Only, previous researchers presented the different coefficient (a, b) of H according to their experimental data. Their coefficients were summarized in Table.2.

$$H = \frac{\cos\theta_S - \cos\theta_A}{1+\cos\theta_S} = aCa^b \qquad (10)$$

In figure.10, Lee and Lee's correlations was well-predicted θ_A of our research in the cases of θ_S =110° and 77°, however other correlations was underestimated θ_A of our research.

Table.2 Coefficients of H in previous θ_A correlations

Researches	Coefficient of H		θ_S range
	a	b	
Jiang et al.	4.96	0.702	All
Bracke et al.	2	0.5	All
Seebergh et al.	2.24	0.54	All
Lee and Lee (1)	0.82	0.118	θ_S =110°
Lee and Lee (2)	2.48	0.328	θ_S =77°

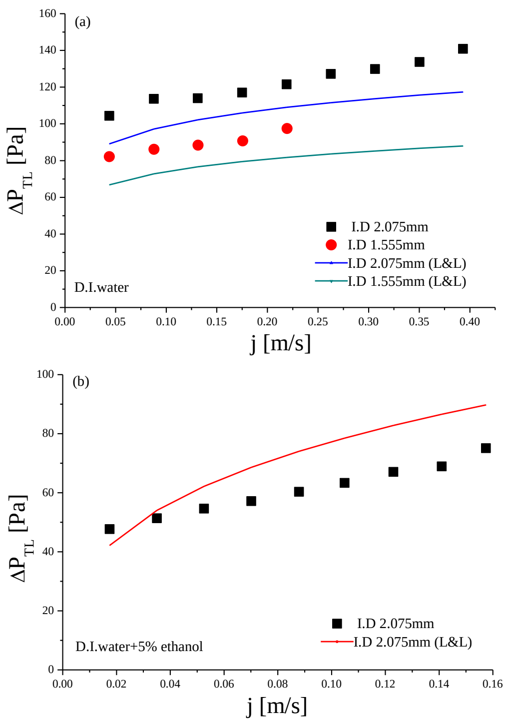

Figure.9 Comparison ΔP_{TL} with Lee and Lee correlation (a) D.I.water, (b) D.I.water+5%ethanol

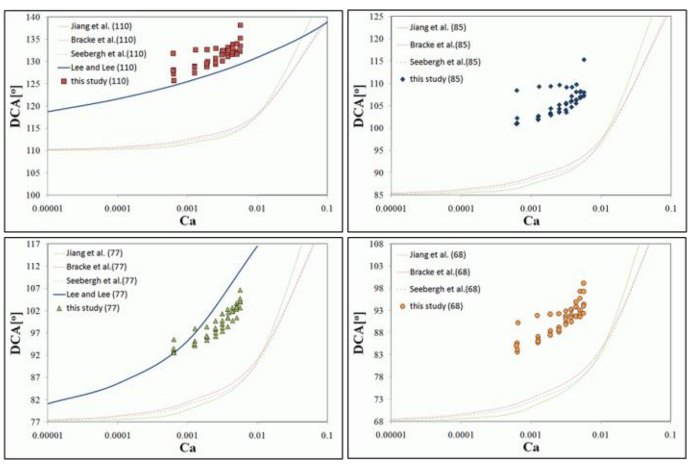

Figure.10 Comparison of θ_A of data and correlations

Table.3 Classification of movement of liquid-gas interface

dominant forces		
interfacial/viscous	interfacial/viscous /gravitational	interfacial/viscous /inertial
Jiang et al.	Bracke et al.	Lee and Lee
	Seebergh et al.	Our research

Also, Lee and Lee correlations and θ_A of our research were dependent of θ_S unlike that other correlations were independent of θ_S.

6. DISCUSSIONS

In previous researches of θ_A, Hoffman developed the curve that θ_A was depended of *Ca* and shift factor. Shift factor was depended of θ_S. Also, he classified movement of liquid-gas interface in horizontal capillary tubes. In [3], the movements were dependent of viscous force, inertial force, interfacial force at the liquid-gas interface and at the liquid-gas-solid junction. Criteria to classify the movements were based on the dimensionless numbers *Ca* and *We*, and he roughly derived that criterion of *We* was 0.015. Because his experiments were carried out over a low range of *We* ($5.4\times10^{-11}\leq We\leq 1.6\times10^{-5}$), the applicability of his correlation was limited to the movement dominated by interfacial and viscous forces. Instead, he conjectured movement of liquid-gas interface in the high range of *We*. According to his conjectures, liquid-gas interface would be disturbed by inertial force and shift factor would be depended not θ_S but θ_A in the high *We* range.

According to Hoffman's criteria, we could classify previous researches. (Table.3) Most of other previous research was conducted using working fluid of viscous oil type, and movement of liquid-gas interface could be nearly influenced on inertial forces. But, Lee and Lee, our research were carried out using working fluid of water type, movement of liquid-gas interface could be dominantly influenced on inertial forces.

Considering Hoffman's conjectures, we could infer the reasons for the difference θ_A between our research and previous correlation. In the high *We* range, interface of liquid-gas could be more convex because, inertial force distorted interface shape. Therefore, θ_A could be higher than predicted θ_A using previous correlation that was developed in the low *We* range.

Also, we could infer the reasons that the coefficients of H were depended of static contact angle. Jiang et al. generated H to take a nonlinear regression Hoffman's curve that θ_A was depended of θ_S and shift factor. Merit of H was that θ_A could be predictable using only θ_A and Ca. Shift factor that a priori was not given was not necessary. H of Jiang et al. was taken a nonlinear regression over a low *We* range. Therefore, it may be that H wasn't well taken a nonlinear regression over a high *We* range.

Eventually, we developed a corrected H over a high We range. The new correlation predicted ΔP_{TL} which agreed with experimentally-measured ΔP_{TL} within 26% and θ_A which agreed with measured θ_A within 5%. This correlation is limited on the conditions $10^{-4}\leq Ca\leq 10^{-3}$, $10^{-2}\leq We\leq 10^{-1}$, $68°\leq \theta_S \leq 110°$

$$H^{*}=\frac{\cos\theta_{S}-\cos\theta_{A}}{\left(1+\cos\theta_{S}\right)^{0.128}}=0.8Ca^{0.399} \tag{10}$$

7. CONCLUSIONS

In this research, we carried up the experimental research about ΔP_{TL} and θ_A. We measured pressure drop of slug with triple-line in a hydrophobic microchannel. To understand the parameters that effected ΔP_{TL}, we experiment in various diameters, fluids and superficial velocities. ΔP_{TL} and θ_A was calculated using measured ΔP_{TP} and equations. Through the experiments, we investigated that ΔP_{TP} is increased as diameter decreased, j* and superficial velocity increased, and this trends are agreement with equations of previous researchers.

Subsequently, ΔP_{TL} and θ_A *of* our research were compared with previous researches. We discovered previous θ_A

correlations were underestimated θ_A of our research. We conjectured the reason of difference with previous θ_A correlations and our research was that previous correlations couldn't consider the inertia effect at the triple-line and interface. Finally, we developed the correlated H over a high *We* range.

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