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CAPILLARITY EFFECT ON TWO-PHASE FLOW RESISTANCE IN MICROCHANNELS

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ABSTRACT

Two-phase flow plays a major role in number of technologies used in fuel cells, bioreactors, phase and particle separators, thermal management systems, chemical reactors, etc. Some of these applications involve air–water two-phase flow in channels much less than 1 mm in diameter. The fundamental understanding of flow characteristics, such as flow regime, pressure drop, and heat transfer, is essential in the design and control of these devices.

In this work, an experimental set-up is developed to investigate the effects of surface energy/surface wettability and geometry on characteristics of two-phase flow in horizontal microchannels at adiabatic conditions. Two-phase (air-water) slug flow is established in microchannel test sections of varying sol-gel dip coated surface wettabilities. Pressure drop measurements and flow pattern detection by high speed visualization are employed to characterize the flow.

Results indicate that two-phase flow resistance is a strong function of surface wettability and it increases with static contact angle. Significant change in contact line in advancing and receding interface, and thus increase in flow resistance, is observed with increase of hydrophobicity while geometry of the channels influences the frequency of the slugs. Presence of thin liquid film, whose thickness is a strong function of wettability, is observed experimentally which clearly elucidates the pressure drop variation with surface wettability. Hence for characterizing two-phase flow in microchannels surface wettability parameters should be accounted appropriately.

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Chapter 1

INTRODUCTION

1.1 Microfluidic Devices

Fluid flow in microchannels has emerged as an important area of research interest due to its seemingly endless and diverse applications in micropower generation, biomedical use, computer chips, chemical separations processes, etc. Advances in microelectro-mechanical systems (MEMS) and other microtechnologies like Micro Direct Methanol Fuel Cell (μ DMFC) involving fluid transport require the use of microfluidic components.

Ashraf et al.¹ summarized the advantages of miniaturizing systems as follows:

- Both chemical and thermal transport processes depend on the surface area of heat exchangers or reactors. As a result, a higher surface-to-volume ratio of a miniature system helps to make a more compact system with a higher volumetric transport coefficient.
- There is increased safety due to its smaller size.
- Miniaturization makes newer engineering systems possible.
- Fourth, there is better cost efficiency.
- Finally, there are more options in usage due to smaller modular size.

Two-phase flow is commonly encountered in many of these microsystems. Some of these applications, like Membrane Electrode Fuel Cells, involve air–water two-phase flow in channels

much less than 1 mm in diameter. As the compactness increases possibly a higher pressure drop is observed in micro- or mini- channels.

It is recognized today that two-phase flow in large channels and pipes is still scientifically one of most challenging problems to be explored since the 1940s, when Lockhart and Martinelli² published a two-phase flow model treating flow as a pseudo-homogenous mixture. At microscale level surface forces become predominant over other body forces. Unlike large-scale systems, gas bubbles can present significant problems in microfluidic systems by disturbing and eventually blocking the flow. Interactions on the boundaries between gas, liquid, and solid introduce nonlinearity and instabilities. Understanding how bubbles affect the flow resistance in microchannels is, besides its fundamental aspect, a concern of determining the pumping or energy requirement for portable microfluidic devices where two-phase flow is involved.

Given that contact angle of liquid (characteristic signature of surface energy) influences the motion of the interface; characteristics of two phase flow in microchannels at varied surface wettabilities are bound to vary from those observed in common microchannels. Gas-liquid interface behavior in micro-channel is more transient and complex than single phase flow and creates many problems related to high surface tension and wetting effects. Interactions on the boundaries between gas, liquid, and solid introduce nonlinearity and instabilities.³ Understanding how surface wettability and geometry of microchannels affect bubbles, thus the flow resistance, in microchannels is a concern of design and process control of these microfluidic devices.

1.2 In this Thesis

In this work, we aim to characterize the effects of surface wettability and geometry on flow parameters (like flow pattern, flow resistance, dynamic contact angle) of air-water two-phase flow in horizontal glass microchannels of various hydraulic diameters at adiabatic conditions. Surface wettability of both circular and square microchannels is varied from superhydrophilic to superhydrophobic by sol-gel dip coating. Pressure drop measurements and flow pattern detection by visualization are employed to characterize the two-phase intermittent slug flow established in microchannels. The results of present study imply that surface wettability and geometry effects are significant on two-phase flow in microchannels and should be accounted appropriately for flow characterization.

Chapter 2

FUNDAMENTAL DEFINITIONS AND FLOW PARAMETERS

2.1 Non-Dimensional Numbers

Hydraulic Diameter (D_h): In order to compare the flows in different geometrical channels, hydraulic diameter is used in characterization of the flows. It is defined as ratio of cross-sectional area of the fluid A to the wetted perimeter P_L .

$$D_h = \frac{4A}{P_L} \quad (1)$$

In the case of completely wetted circular tubes, D_h = Diameter of tube

Reynolds number (Re): It represents the ratio of inertia forces to the viscous forces.

$$Re = \frac{\rho UL}{\mu} \quad (2)$$

where L is the characteristic length of the flow scale. For microchannels of various geometries, D_h is used as characteristic length.

Weber Number (We): Liquid Weber number represents the ratio of inertia to surface forces

$$We = \frac{\rho U^2 D}{\sigma} \quad (3)$$

Capillary Number (Ca): It represents the relative effect of viscous forces versus surface tension acting across an interface between a liquid and a gas, or between two immiscible liquids.

$$We = \frac{\mu U}{\sigma} \quad (4)$$

Bond Number (Bo): It represents the ratio of body forces (often gravitational forces) to surface tension forces.

$$Bo = \frac{\rho g L^2}{\sigma} \quad (5)$$

Suratman Number (Su): Also known as Laplace number (La), it is a dimensionless number used in the characterization of free surface fluid dynamics. It represents ratio of surface tension to the momentum-transport (especially dissipation) inside a fluid.

$$Su = La = \frac{\sigma \rho L}{\mu^2} \quad (6)$$

2.2 Microchannels

Microchannels can endure a higher operating pressure and provide a much larger contact area with fluid per unit volume than large tubes. The high degree of compactness yields new application areas for such devices, which increase as they advance to smaller sizes.

Flows in microchannels are addressed by Gad-el-Hak⁴ in micro-electromechanical devices with a characteristic size ranging from 1 mm down to 1 μm . The behavior of such flows differs from those in macroscopic channels basically in the following three features:

- The Reynolds number is lower and therefore the flow is laminar or even of Stokes type, i.e., without inertia. Care is needed, however, in applying the well-known macroscopic laws to flow in a microchannel. An earlier onset of transition to turbulent flow at $Re \approx 2000$ is observed.
- Averaging over characteristic lengths gives rise to microscopic effects which invalidate the continuum approach in the limit of molecular length scales.
- Capillary forces influence or even dominate the flow while gravity effects can be neglected.

2.2.1 Macro-micro Transition

A tricky aspect in micro-scale studies is - how to identify the macro-to-micro scale threshold. Many methods⁵⁻¹⁰ are adopted for the conventional-to-micro channel threshold based on the characteristic channel cross-section dimensions. One classification based on fixed channel hydraulic diameters was also proposed by Mehendal⁴ as follows: microchannels (1–100 μm), mesochannels (100 μm –1 mm), macrochannels (1–6 mm) and conventional channels ($D_h > 6$ mm). Due to many methods and lack of physical significance, it is still far from clear as how to define the macro-to-micro transition.

In this study, hydraulic diameters larger or equal to threshold diameter (D_{th}), suggested by Triplett⁸, and those smaller than D_{th} were denominated as macrochannels and microchannels, respectively.

$$D_{th} = L = \sqrt{\frac{\sigma}{(\rho_L - \rho_G)g}} \quad (7)$$

2.3 Two-phase Flow

Ink jet printing was probably the first microfluidic application involving two-phase flow. As capillary gas-liquid two-phase flow occurs in increasingly more modern industrial applications like thermal management systems, fuel cells, etc., there has been a notable growth in the number of studies on two-phase flow and evaporation heat transfer in micro-scale channels in recent years.

Classical Thermodynamics states that phase is a macroscopic state of the matter which is homogeneous in chemical composition and physical structure; e.g. a gas, liquid or solid of pure component. Two-phase flow is the simplest multiphase flow in which two phases are present for a pure component. The two fluid phases can be both liquids, such as water, oil, paint or lubricant, or a liquid and a gas –vapor phase.

2.3.1 Contact Line

When two immiscible fluids are placed in a container or on a substrate, the line where the interface of the two fluid phases intersects the solid container or the substrate is called the *contact line*, as shown in Fig. 2.3.1. The equilibrium configuration of the static contact line was the topic of classical work of Young, Laplace, and Gauss¹¹, and is described by Young's equation which relates the three coefficients of interfacial tension to the contact angle formed by the fluid-fluid interface with the solid surface.

Fig. 2.3.1 Contact line of gas-liquid interface of two-phase flow in microchannel

2.3.2 Two-phase Flow Pattern

Distribution of the two fluids inside the flow channel is called as *flow pattern* or *flow regime*. When a gas and a liquid are forced to flow together inside a pipe, there are many different flow regimes, which occur for flow in macro- and micro- channels. The regime depends on the fluid properties, the size of the conduit and the flow rates of each of the phases. The flow regime can also depend on the configuration of the inlet; the flow regime may take some distance to develop and it can change with distance as (perhaps) the pressure, which affects the gas density, changes. For fixed fluid properties and conduit, the flow rate is the independent variable which influences changes in the flow regime.

Flow patterns commonly observed in the adiabatic two-phase flow can be categorized into: Bubbly, Plug, Slug, Stratified, Annular, and Rivulet. Generally observed flow patterns are shown in Fig. 2.3.2 and are described in brief as follows:

Bubbly Flow: In this the liquid flow rate is high enough to break up the gas into bubbles, but it is not high enough to cause the bubbles to become mixed well within the liquid phase.

Plug Flow: It is characterized by a continuous stream of vapor plugs flowing in the liquid. A thin film of liquid coats the tube wall and surrounds the vapor plug. Small disturbances may exist at the advancing and receding interfaces of the bubbles but as a whole the plugs remain intact and uniform.

Slug Flow: As the gas mass flow rate is increased for plug flow, the disturbances amplify until the receding portion of the plug breaks apart into smaller bubbles. At this point the flow pattern becomes slug flow. These smaller bubbles become trapped in the liquid flow and impact the front of the following slug causing disturbances in the front of the flow profile.

Stratified Flow: The two phases are separated with all liquid flowing at the bottom of the channel (under normal gravity conditions). This flow pattern occurs at low liquid and gas velocities.

Wavy Flow: As the gas velocity is increased, waves are formed on the liquid-gas interface, with the waves travelling in the direction of the flow. This flow pattern corresponds to stratified wavy flow.

Annular Flow: This flow regime consists of a nearly complete separation of the liquid and gas along the circumference of the tube wall. The first form of annular flow occurs when the surfaces of waves in wavy flow amplify to the extent that they touch the top of the tube wall. This flow pattern is known as wavy annular flow “pseudo slug flow”. As the mass flow rate is increased the liquid is pushed up around the circumference of the tube wall by the increase in the gas momentum and falls downward under gravity in the form of annular waves. When the liquid coats the tube wall completely forming an annular ring of the liquid phase and the gas flows through the core of the tube, the flow pattern is known as annular flow. In annular flow there could also be small droplets entrained in the gas core. This flow is also known as annular mist flow.

Many researchers¹²⁻¹⁵ have described the importance of using flow pattern information in the determination of accurate two-phase flow models. Consequently, much attention has been directed towards developing two-phase flow regime maps. There are three main types of two-phase flow maps in the literature: Baker/Mandhane type, Taitel–Dukler type, and Steiner type. Flow regime maps are developed by using superficial gas and liquid velocities and some fluid property scaling factor. Most of the recent two-phase flow regime maps found in literature is of Steiner type flow maps with quality on the horizontal axis and mass flux on the vertical axis.

(a) Bubbly Flow	(b) Slug Flow
(c) Transition	(d) Skewed Flow (Yakitori Flow)
(e) Liquid Ring Flow	(f) Frothy Annular Flow
(g) Transition	(h) Annular Flow
(i) Rivulet Flow	

Fig. 2.3.2 Commonly observed Two-phase flow patterns for flows in macro- and micro-channels

2.3.3 *Two-phase Flow Transitions*

Similar to the classic Moody diagram in single-phase flow, as per Revellin et al.¹⁶, three zones are distinguishable when plotting the variation of the two-phase friction factor versus the two-phase Reynolds number: a laminar regime for $Re_{TP} < 2000$, a transition regime for $2000 < Re_{TP} < 8000$ and a turbulent regime for $Re_{TP} > 8000$.

2.3.4 *Two-phase Flow Resistance (Pressure Drop)*

It has long been known that the pressure drop for two phase flow is much greater than that of a single phase fluid. While the introduction of a second phase results in a reduction of the flow area of both phases this alone cannot account for the dramatic increase in pressure drop. Boelter and Kepner¹⁷ showed that the introduction of a small amount of liquid into the gas flow increased the pressure drop by 4 times. This small amount of liquid is pushed either to the bottom or around the circumference of the pipe and ripples are formed by the gas pushing on the liquid. The result is a large loss of energy in the gas phase.

In comparison to single-phase flow, e.g., of water or air alone, two-phase flow is significantly different due to the presence of other independent variables such as velocity of the mixture and its components, spatial concentration, film thickness, and the spatial and temporal distributions of these parameters. The effect of these additional variables creates interesting challenges in describing the flow phenomena, since these variables greatly influence the nature of the two-phase flow, resulting in significant differences in reported results, compounded by difficulty in defining the experimental conditions, in verification and validation of measured and calculated data, and in comparing the results of different experimenters.

The total pressure drop of a fluid is due to the variation of kinetic and potential energy and that due to friction, so that the pressure drop is the sum of the static pressure drop (elevation head), the momentum pressure drop (acceleration) and frictional pressure drop¹⁶:

$$\left(\frac{dP}{dz} \right)_t = \left(\frac{dP}{dz} \right)_s + \left(\frac{dP}{dz} \right)_f + \left(\frac{dP}{dz} \right)_f \quad (8)$$

The static pressure drop in a horizontal microchannel is 0.

$$\left(\frac{dP}{dz} \right)_s = 0 \quad (9)$$

The momentum pressure drop takes into account the acceleration of the flow due to the flashing or diabatic effect and is defined as follows:

$$\left(\frac{dP}{dz} \right)_t = G^2 \frac{\Delta x (\rho_L - \rho_G)}{L \rho_L \rho_G} \quad (10)$$

The two main approaches used to model frictional pressure gradient $(dP/dz)_f$ in macro- and micro-scale two-phase gas–liquid flow are the “homogenous” model and the “separated” flow model. In the first model, both liquid and vapor phases move at the same velocity (slip ratio = 1). Consequently, the homogeneous model has also been called the zero-slip model. The homogenous model, which is the simplest one, is based on the assumption that the two-phase fluid behaves as a pseudo-single-phase fluid presenting pseudo-properties that are weighted relative to the vapor and liquid flow fraction. Different ways to define the pseudo-properties,

usually called mixture-properties, have been proposed that are well detailed in Collier and Thome¹⁸. Thus, the frictional pressure drop is calculated by assuming a constant friction coefficient between the inlet and outlet sections.

In the ‘separated flow model’, two-phase flow is considered to be divided into liquid and gas streams, each flowing in its own pipe. Hence, the separated flow model has been referred to as the slip-flow model. The cross-sectional flow areas of the two pipes are proportional to the void fraction. The basic equations for the separated flow model are not dependent on the particular flow configuration adopted. It is assumed that the velocities of each phase are uniform, in any given cross-section, within the zone occupied by the phase. The separated flow model has been continuously developed since 1949 when Lockhart and Martinelli² published their classic paper on two-phase gas liquid flow. The main goal in this approach is to find an empirical correlation or simplified concept to relate the two-phase friction multiplier, to the independent variables of the flow.

Both the homogeneous and separated flow type models are often used in all ranges of qualities and mass fluxes. For some of the situations the assumptions of physics of the flow does not hold good. However, more and more experimenters are using in situ parameters in their investigations.

In two-phase micro-scale flows, the liquid phase is laminar for almost all the test conditions, which is quite rare in macro-scale studies. In addition, it is expected in small diameter tubes that the effect of surface tension may become more pronounced while the influence of gravity may become less important and consequently stratified flows are rarely observed. Bubbly flow is also seldom observed due to the fact that its lifespan is very short as bubbles coalesce or grow to the channel size very quickly. Thus, the correlations developed for macro-scale may not extrapolate

well to the micro-scale and specific correlations have been proposed to predict pressure drop of two-phase flow in microchannels.

Furthermore, from an overall point of view, none of the present available methods can be classified as a general design method since even the best ones are only able to capture about one-half the data to within $\pm 30\%$.

2.4 Surface Tension / Surface Energy /Surface Wettability.

The laws of thermodynamics state that as system seeks equilibrium it will obtain lowest available energy state. This of course applies to liquids and their interfacial relation with solid surfaces. For example, a drop of water placed on a planar surface acquires a partial spherical shape due to the interfacial surface tension forces acting on it. Interfacial surface tension comes into play because of various molecular interactions at the interface. For instance, water molecules are highly polar, therefore strongly self-attractive, and water has a high surface tension of ~ 73 mN/m.

Surface tension can be expressed as the work done per unit increase in surface area, as given $\gamma = dW/dA$, where γ (N/m) is the surface tension, dW is the change in energy or work done, dA is the change in surface area. A classic example is the work required to pull a soap film across a wire loop of given area. Water is one of the most polar liquids at ~ 73 mN/m.

Ability of fluid to spread on the surface is known as *Surface wettability*.

2.4.1 Static Contact Angle

Fig. 2.4.1. Liquid droplet on a solid surface. Interfacial surface tensions at the solid-gas (γ_{SG}), solid-liquid (γ_{SL}) and liquid-gas (γ_{LG}) interface are shown

As shown in Fig. 2.4.1., liquid droplet placed on a surface attains a spherical shape which allows it to minimize its surface area. The droplet contact angle (θ_C) for such a three phase solid -liquid - gas system is given by the well known Young's equation,

$$\cos \theta_C = \frac{\gamma_{SG} - \gamma_{SL}}{\gamma_{LG}} \quad (11)$$

Here, the interfacial surface tension forces balance at the vertex of the 3-phase system. This vertex is often referred to as the liquid contact line or triple-point. A larger Young's contact angle θ_C corresponds to a larger solid-liquid interfacial surface tension.

Contact angle (CA), which is defined by Young's equation¹¹, is the characteristic signature of the surface wettability / surface energy. Low CA represents high surface wettability (i.e. low surface energy) while higher CA represent less surface wettability (i.e. high surface energy). Adding surfactants to the fluids alters the surface tension (and contact angle) and eventually the thermo-physical properties of fluid too.

2.4.2 *Dynamic and Apparent Contact Angle*

Dynamic Contact Angle (θ_d) is the angle, calculated through liquid phase as shown in Fig. 2.4.2, which the moving gas-liquid interface makes with the surface of the wall.

Fig. 2.4.2 Dynamic contact angle (θ_d) of moving air-water interface in contact with solid wall

Apparent Contact Angle (θ_a) is the maximum value of the angle along the interface i.e. maximum value of dynamic contact angle.

Chapter 3

LITERATURE REVIEW AND PROBLEM STATEMENT

In this section, we present the literature relevant to the present investigation of capillarity effects on two-phase flow in microchannels.

3.1 Adiabatic Two-phase Flow in Microchannels

The studies found from a literature search on frictional pressure drop in micro-scale channels revealed that experiments were performed under both diabatic and adiabatic conditions, for air–water mixtures and halocarbon refrigerants. Only studies related to adiabatic two-phase flow in microchannels are listed down in Table 1, which includes the description of the test geometries and experimental conditions.

There are many publications dealing with the two-phase flow characteristics in capillary tubes with a hydraulic diameter of mini and microchannels. (Revellin and Thome¹⁶, Son and Allen^{20,21}, Bi and Zhao^{22,23}, Lee and Lee²⁴, Yang and Shieh²⁵, Chen et al.²⁶, Kawaji et al.²⁷⁻²⁹, Triplett et al.⁸, Coleman and Garimella^{12,14,30}, Mishima and Hibiki³¹, Keska et al.^{32,33}, Fukano and Kariyasaki³⁴, Nino et al.³⁵, Barajas and Panton³⁶, Damianides and Westwater³⁷, Suo and Griffith³⁸, Hidrovo et al.^{19,39}, Serizawa et al.⁴⁰; among others).

Table 1- Review of studies considering surface tension / surface wettability effects on adiabatic two-phase flow

Authors	Geometry / Orientation	Channel material	D_h / L (mm)	Fluid	Experimental condition
Suo & Griffith ³⁸	Circular / Horizontal & Vertical	Glass	1.0–1.6 / -	Water-air Water-N ₂	Adiabatic
Damianides and Westwater ³⁷	Circular	Pyrex glass	1.0-5.0 / -	Water-air	Adiabatic
Barajas and Panton ³⁶	Circular / Horizontal	Pyrex, polyethylene, polyurethane, FEP	1.6 / 300	Water-air	Adiabatic
Son & Allen ^{20, 21}	Circular ^a , Rectangular ^b / Horizontal	Quartz	0.33 ^a , 0.50 ^b / 127 ^a , 102 ^b	Water – Nitrogen	Adiabatic
Keska et al. ^{32,33}	Square / Horizontal	Glass	0.996/105	Water-air	Adiabatic
Mishima and Hibiki ³¹	Circular / Vertical	Glass, aluminum	1.05-4.08 / 210-1000	Water-air	Adiabatic
Triplett et al. ⁸	Circular ^a , triangular ^b / Horizontal	Ace glass, Pyrex	1.1 ^a , 1.45 ^a , 1.09 ^b , 1.49 ^b / 200	Water-air	Adiabatic
Nino et al. ³⁵	Rectangular / Horizontal	-	1.54&1.02 / 610 & 1070	R134a, R410A, N ₂ , Air-Water	Adiabatic
Kawaji et al. ²⁷⁻²⁹	Circular / Horizontal	Fused silica	0.05/46 0.10/64,65 0.25/157 0.53/277	Water – N ₂	Adiabatic
Serizawa et al. ⁴⁰	Circular / Horizontal	Quartz	0.01 – 0.1 / 10-14	Water – air Water-steam	Adiabatic

Mishima and Hibiki³¹ studied flow regime, void fraction, rise velocity of slug bubbles, and frictional pressure loss for air-water flows in vertical capillary tubes with inner diameters in the range of 1 to 4 mm. The results were compared with the Lockhart and Martinelli model. The frictional pressure loss was reproduced well by Chisholm's equation with a new equation for Chisholm's parameter C as a function of inner diameter. Their correlation was

$$C = 21 (1 - e^{-319 D_h})$$

Triplett et al.⁸ investigated experimentally the void fraction and two-phase frictional pressure drop in microchannels. They used an air-water mixture as a working fluid. They carried their experiments in transparent circular microchannels with $d = 1.1$ and 1.45 mm and in microchannels of semi-triangular (triangular with one corner smoothed) cross-sections with $D_h = 1.09$ and 1.49 mm. They varied the superficial velocity of gas and liquid phases in the range of $U_g = 0.02$ – 80 m/s and $U_l = 0.02$ – 8 m/s, respectively. They calculated void fractions by analyzing photographs taken from the test sections of circular cross-section. Based on the numerical solution of mass and momentum conservation equations, they applied a one dimensional model for the calculation of test section pressure drops using different two-phase friction models. For bubbly and slug flow patterns, they found that the two-phase friction factor based on homogeneous mixture assumption provided the best agreement with experimental data. For annular flow, they found that the homogeneous mixture model and other widely used correlations significantly over-predicted the frictional pressure drop.

Kawaji et al.²⁷⁻²⁹ conducted an experimental investigation on two-phase flow characteristics in a circular tube of $d = 50$ μm to 250 μm . They used de-ionized water-nitrogen gas mixture as a working fluid and determined two-phase flow patterns by video recording the flow in the transparent capillary tube made of fused silica. They varied the superficial velocity of gas and

liquid phases in the range of $U_g = 0.1\text{--}60$ m/s and $U_l = 0.02\text{--}4$ m/s, respectively. They obtained and analyzed time-averaged void fraction and two-phase friction pressure drop data. They found that the observed flow patterns were intermittent and semi-annular flows, but a closer study of the liquid film structure revealed gas core flows with a smooth or ring-shaped film and a serpentine like gas core surrounded by a deformed liquid film. They did not observe bubbly and churn flow patterns. Based on the probability of the appearance of each type of flow, they developed a flow pattern map and compared it to the existing flow pattern maps obtained for 1 mm diameter channels. Compared to the flow in channels with $D_h = 1$ mm, they found that the time-averaged void fraction remained low even at high gas flow rates, indicating significantly larger slip ratios and weaker momentum coupling between the phases. They showed that the single-phase friction factor and two-phase friction multiplier data agreed well with the conventional correlations.

In another separate study, they investigated experimentally the effect of channel diameter on two-phase flow to identify the phenomena that distinguish microchannels from minichannels. They used nitrogen gas-water mixture as a working fluid. They carried their experiments in circular channels of $d = 530, 250, 100,$ and 50 μm ; measured the temperature, pressure, and flow rates of the liquid and gas phases; and recorded the images of the flow patterns. They characterized two-phase flow by the flow regime map, void fraction, and frictional pressure drop, and found that the two-phase flow characteristics in the 530 and 250 μm channels were similar to those typically observed in minichannels of 1 mm diameter, while the two-phase flow behavior in the 100 and 50 μm channels departed from that observed in minichannels. In the 100 and 50 μm channels, the slug flow is dominant. The void fraction-volumetric quality relationship departed from a linear Armand-type correlation, and mass flux no longer influenced the two-

phase frictional multiplier. In the range of channel diameters investigated, they showed that the channel diameter had a clear effect on two-phase flow. Also, they proposed a new slug flow model to gain physical insight into the observed flow characteristics in microchannels. Their model could predict the two-phase frictional pressure gradient for the 100 and 50 μm channels using actual void fraction data, substantiating the assumed flow mechanism.

Serizawa and Feng⁴⁰ conducted air–water and steam-water two-phase flow experiments in 25 and 50 μm capillary tubes and reported the observation of a liquid ring flow pattern, unique to flow channels of these dimensions.

Son and Allen²⁰⁻²¹ visualized the effect of wettability on microchannel two-phase flow resistance. They illustrated that uncoated microchannels (hydrophilic in nature) show stable flow with high velocity liquid slugs translating on an annular liquid film while a commercial polymer coated microchannel (hydrophobic in nature) exhibits less stable flow with droplet formation and contact line drag.

3.2 Pressure Drop of Two-phase Flow in Microchannels

Actually, the two-phase friction takes into account the friction between the liquid and the wall as well as the friction between the liquid and the vapor, the latter being much higher in case of wavy-annular regime. Table 2 below lists the models which are widely used for microchannels.

Lee and Lee²⁴ proposed new correlations for the two-phase pressure drop through horizontal rectangular channels with small gaps (heights) based on 305 data points. The gap between the upper and the lower plates of each channel ranges from 0.4 to 4 mm while the channel width was fixed to 20 mm. Water and air were used as the test fluids. The authors expressed the two-phase

frictional multiplier using the Lockhart–Martinelli type correlation but with the modification on parameter C, see Table 3.2. (b).

Table 2- Pressure drop models and correlations for microchannels

Pressure drop models and correlations for microchannels	
Authors	Equation
Mishima and Hibiki [31]	$C = 21(1 - e^{-3.19D})$ using the Lockhart–Martinelli model
Lee and Lee [24]	$C = A\lambda^q\psi^r Re_{LO}^s$ using the Lockhart–Martinelli model $\lambda = \frac{\mu_L^2}{\rho_L \sigma D}$ and $\psi = \frac{\mu_j}{\sigma}$ The exponents A , q , r and s are given in Table 3
Lee and Mudawar [41]	For laminar liquid and laminar vapor: $C_{vv} = 2.16 Re_{LO}^{0.047} We_{LO}^{0.6}$ For laminar liquid and turbulent vapor: $C_{vt} = 1.45 Re_{LO}^{0.25} We_{LO}^{0.23}$ with $We_{LO} = \frac{G^2 D}{\rho_L \sigma}$ using the Lockhart–Martinelli model
Zhang and Webb [42]	$\left(\frac{dP}{dz}\right)_f = \left(\frac{dP}{dz}\right)_{LO} \Phi_{LO}^2$ with $\Phi_{LO}^2 = (1 - x)^2 + 2.87x^2 \left(\frac{P}{P_{crit}}\right)^{-1} + 1.68x^{0.8}(1 - x)^{0.25} \left(\frac{P}{P_{crit}}\right)^{-1.64}$
Tran et al. [43]	$\left(\frac{dP}{dz}\right)_f = \left(\frac{dP}{dz}\right)_{LO} \Phi_{LO}^2$ with $\Phi_{LO}^2 = 1 + (4.3Y^2 - 1)$ $\left[Co x^{0.875}(1 - x)^{0.875} + x^{1.75}\right]$ $Co = \left(\frac{\sigma}{D^2 g(\rho_L - \rho_V)}\right)^{0.5}$ and $Y^2 = \frac{(dP/dz)_{VO}}{(dP/dz)_{LO}}$

Table 3- Components and exponents for parameter C of Lee and Lee⁴¹

Constant and exponents for parameter C of Lee and Lee [6]								
Flow regime liquid	Gas	A	q	R	S	Range of X	Range of Re_{LO}	Number of data
Laminar	Laminar	6.833×10^{-8}	-1.317	0.719	0.557	0.776–14.176	175–1480	106
Laminar	Turbulent	6.185×10^{-2}	0	0	0.726	0.303–1.426	293–1506	2
Turbulent	Laminar	3.627	0	0	0.174	3.276–79.415	2606–17,642	85
Turbulent	Turbulent	0.408	0	0	0.451	1.309–14.781	2675–17,757	62

Laminar: $Re_L, Re_G < 2000$; turbulent: $Re_L, Re_G > 2000$.

Awad and Muzychka⁴⁴ proposed simple expressions for obtaining bounds for two-phase frictional pressure gradient in minichannels and microchannels. The lower bound is based on Ali et al. correlation for laminar-laminar flow. This correlation is based on the modification of a simplified, stratified flow model derived from the theoretical approach of Taitel and Dukler for the case of two-phase flow in a narrow channel. The upper bound is based on Chisholm correlation for laminar-laminar flow. The mean model is based on the arithmetic mean of lower bound and upper bound. Their model is verified using published experimental data of two-phase frictional pressure gradient in circular and non-circular shapes, and concluded that proposed model bounds the published data well for a wide range of mass fluxes, mass qualities, diameters, and saturation temperatures.

The physical meaning of the lower bound ($C = 0$) is that the two-phase frictional pressure gradient is the sum of the frictional pressure of liquid phase alone and the frictional pressure of gas phase alone:

$$\left(\frac{dp}{dz}\right)_{f,lower} = \frac{32G(1-x)\mu_l}{d^2\rho_l} * \left[1 + \left(\frac{x}{1-x}\right) \left(\frac{\rho_l}{\rho_g}\right) \left(\frac{\mu_g}{\mu_l}\right)\right] \quad (12)$$

$$\begin{aligned} \left(\frac{dp}{dz}\right)_{f,upper} &= \frac{32G(1-x)\mu_l}{d^2\rho_l} \\ &* \left[1 + 5 \left(\frac{x}{1-x}\right)^{0.5} \left(\frac{\rho_l}{\rho_g}\right)^{0.5} \left(\frac{\mu_g}{\mu_l}\right)^{0.5} \right. \\ &\quad \left. + \left(\frac{x}{1-x}\right) \left(\frac{\rho_l}{\rho_g}\right) \left(\frac{\mu_g}{\mu_l}\right)\right] \end{aligned} \quad (13)$$

This model can be applied for circular shapes using tube diameter, d , as well as the hydraulic diameter, D_h . For non-circular shapes, the Hagen-Poiseuille constant = 16 in Eq. (12) will be changed. For example, for a rectangular channel with the aspect ratio of 0, the Hagen-Poiseuille constant = 24, while for a rectangular channel with the aspect ratio of 1 (square channel), the Hagen-Poiseuille constant = 14.23.

Garimella et al.³⁰ proposed the first mechanistic model for two-phase pressure drop during intermittent flow of refrigerant in circular microchannel. The model was developed for a unit cell in the channel based on the observed slug/bubble flow pattern for these conditions. The unit cell comprises a liquid slug followed by a vapor bubble that is surrounded by a thin, annular liquid film. Authors first of all accounted for the continual uptake of liquid from film into the front of the slug ($\Delta P_{\text{film_slug}}$ transitions) as the bubble travels faster than the liquid slug. They also accounted for the pressure drop in bubble/film interface ($DP_{f/b}$) and the pressure drop in the liquid slug (ΔP_{slug}). An experimental correlation is proposed for predicting the slug frequency. It was assumed in the model that the length/frequency/speed of bubble/slug is constant, with no bubble coalescence and a smooth bubble/film interface. The model has been experimentally validated for condensing R-134a.

3.3 Effect of Surface Wettability on Two-phase Flow Bubble Dynamics

As surface area to volume ratio is very high for microchannels, surface forces become predominant over body forces for microscale applications. At microfluidic scales, the behavior of liquid film plays a critical role in flow regime transition. Many studies (like Cubaud⁴⁵) have reported that the contact angle (surface wettability) is an important consideration in the design of micro, and even more so, nano fluidic devices. Rosengarten et al.⁴⁶, have reported slip flow that

is known to occur more readily on hydrophobic surfaces and above a critical shear rate. For fully wetting surfaces (a contact angle of zero) one would not expect slip and thus the gradient should be zero. While most of the two-phase flow literature reports experiments conducted with a complete wetting fluid, wetting often takes place either on heterogeneous surfaces or with contaminant preventing a complete wetting of the system and leading to the formation of a contact line. Contact line dynamics along with the role of liquid film stability where phase interaction is a combination of viscous and capillary effects are shown to be a more appropriate scaling for two-phase flows.

While capillarity has been studied extensively, capillarity in microchannels has not received much attention. For annular films without an imposed axial flow and no surface shear, it has been observed that core flow will be pinched off with liquid accumulating into lobes and into lenticular lenses, if the volume of liquid is sufficient. The formation of a lenticular lens from the liquid in the annular film is also referred to as “snap-off” or “liquid hold-up”. Whether the adjacent lobes/lenses are connected to one another by a liquid film depends upon the static contact angle between the liquid and the tube and upon van der Waals forces. Thus, the stability of the liquid film can significantly influence the inertia of the system through the morphological change from thin film to plug.

The pinchoff results from a capillary-driven instability; commonly referred to as Rayleigh instability. Many low-Bo studies have shown both experimentally and theoretically, that two-phase flow in straight channels can rapidly undergo liquid-gas configurations changes; slug-bubbly transitions in high-Su flow and liquid lobe/lens formation (pinch-off) in low-Su flow. The liquid-gas configuration changes have been extensively studied for low-Su flows and, thus, provide a solid starting point for examining high-Su flows which have not been studied from a

fundamental physics of fluids viewpoint. Understanding bubble dynamics and thin fluid film dynamics is critical to the design and optimization of microfluidic devices using two-phase flows in microchannels. Son and Allen^{20,21} have reported that as surface wettability is varied, the fluid wicking in to and out of the corner of square is observed.

The majority of the effort in the low-Bo, high-Su (inertial) two-phase flow research has been in determining flow transition in straight, circular tubes. In all these fundamental studies, the importance of the dynamics and stability of liquid films on the behavior of two-phase flow is established over and over again. It is critical to include the physics of the liquid films in scale modeling in order to properly characterize two-phase flow in microchannels.

3.4 Effect of Geometry and Surface Wettability on Two-phase Flow Characteristics

Many researchers have reported specifically the effects of surface tension, tube diameter and aspect ratio on flow pattern transitions.

Suo & Griffith³⁸ performed studies of elongated bubble flow pattern in capillary tubes with diameters ranging from 1.0 to 1.6 mm. They concluded that since capillary slug flow exists in both horizontal and vertical tubes, the surface tension forces dominate over the gravity forces. Damianides & Westwater³⁶ studied two-phase flow in tubes ranging from 1 to 5 mm. They concluded that the surface tension between the gas and liquid is a very important variable for air-water two-phase flow in pyrex glass tube diameters of ≤ 5 mm. Graska⁴⁷, using liquids with different surface tensions, concluded that the liquid-gas surface tension is a significant variable in two-phase flow in pyrex glass tubes of ID ≤ 5 mm. Barajas and Panton³⁶ studied the effects of varied contact angle on the flow in 1.6 mm circular tube and concluded it is an important parameter affecting two-phase flow in capillaries.

Studies by Garimella^{12,30} on two-phase flow implied that for tubes with $d < 10\text{mm}$; aspect ratio, hydraulic diameter and surface tension will be important factors in determining the locations of flow regime transitions.

Due to the capillary force, Bi and Zhao^{22,23} argued that water would always be retained in the corner of the channel and creates the buoyancy force necessary for air bubbles to rise in stagnant water. They had documented the motion of Taylor bubbles in triangular and rectangular channels with a hydraulic diameter as small as D_h 50.866 μm and D_h 51.0 μm , respectively. Carey⁴⁸ described an alternating sequence of liquid and gas slugs in a small circular channel. The gas bubble fills the entire cross section of the tube and yields a dry wall. There is a complete separation of the gas phase from the liquid phase, in contrast to that intrinsic to small noncircular channels. Furthermore, Zhao and Bi stated that the flow in the corner of a triangular channel may stay laminar despite a fully turbulent flow in the core. According to Kolb and Cerro⁴⁹, the shape of a gas bubble inside a square channel depends on the magnitude of the capillary number, Ca . For a large Ca , the cross section of the bubble is axisymmetric and circular. For a small Ca , the bubble shape is non-axisymmetric. Here, the cross section of the bubble appears flat alongside the channel walls and displays an arc of constant curvature in the corners of the square channel. The non-axisymmetric shape of the bubble in horizontal flow is attributed to the dominating effect of gravity at low Ca . Later, Kolb and Cerro claimed that the flow in the corners contributes to over 95% of the total flow. Clearly, the differences between the flows in noncircular and circular channels can be significant due to the flow at the corners.

Son and Allen^{20,21} visualized the effect of wettability on microchannel two-phase flow resistance. They illustrated that uncoated microchannels (hydrophilic in nature) show stable flow with high velocity liquid slugs translating on an annular liquid film while a commercial polymer

coated microchannel (hydrophobic in nature) exhibits less stable flow with droplet formation and contact line drag.

3.5 Problem Statement

As evident from above studies, researchers have ascertained influence of surface tension on flow characteristics by considering fluids of different surface energy. Since fluids with various surface tension have different thermo physical properties, scaling of the results for air-water two-phase flow may not be successful. Very little research has been conducted in non-circular channels. Very few experimental studies are available reporting the effect of surface wettability (contact angle) for air-water adiabatic two-phase flow in horizontal microchannels less than 1mm diameter. As we aim to increase our understanding of air-water two-phase flow, surface wettability of various geometric microchannels are modified. Quantifying the flow physics of pipe and channel flows is thus critical in understanding the momentum, heat, and mass transport characteristics at the microscale.

Chapter 4

EXPERIMENTAL SETUP AND PROCEDURE

In this section, we will discuss surface energy modification procedures, experimental apparatus & procedure, methodology used for analysis in this study.

4.1 Surface Wettability Modification

Surface wettability is described by a contact angle (CA), which is defined by Young's equation [11]. Adding surfactants to the fluids alters the surface tension (and contact angle), but eventually the thermo-physical properties of fluids also. In order to study only air-water two-phase flows in various surface wettability configurations, surface energy of the microchannels is modified accordingly. Further the transparency of the glass microchannels is of utmost importance to enable flow visualization. As the plain glass channels were hydrophilic in nature (contact angle $CA \sim 60^\circ$), we decided for coatings of superhydrophilic ($CA 20^\circ$), hydrophobic ($CA 105^\circ$) and superhydrophobic ($CA 150^\circ$) to obtain entire spectrum of possible wettabilities.

4.1.1 Literature Review: Surface Wettability Coatings

For a given system, there are two basic approaches to modify the contact angle. One is to chemically change the surface chemistry while the other is to change the surface roughness. In this study, both the approaches are used to create the thin films of various surface wettabilities with high optical transparency.

Various methods like Vapor deposition, Chemical Vapor deposition, Plasma deposition, Laser and photo induced, Sol-gel dip coating are available to realize surface coatings. As recessed inner areas of the microchannels are required to be modified for this study, many of the above listed methods are not applicable. On literature review, sol-gel dip coating method is found as most suitable method. In this method, sol flows into the recessed areas of microchannels under capillary action and depending on the withdrawal speed it gives uniform thickness coatings.

Further the sol-gel dip coating method produces highly transparent coatings. It is fast, economical, and efficient surface finishing method with fairly good results. Also it is widely used in automobile glass industry, etc.

4.1.2 Thin Film Coatings

We carried out the coatings of the microchannels so as to obtain superhydrophilic⁵⁰ (CA 15-25°), hydrophobic⁵¹ (CA 100-110°) and superhydrophobic⁵⁰ (CA 135°) surfaces while the plain microchannels were hydrophilic (CA 50-60°) in nature. Since the static contact angle cannot be measured inside the microchannel, a reference section of thin glass substrates is surface treated concurrently with the microchannels to ascertain the contact angle of the coating. Flat plain glass substrates of size 20 mm X 20 mm approx. (obtained from commercially available Corning glass micro slides, No: 2947- 75x50), are coated in the solution along with microtubes and are used for measuring contact angle of thin films. Glass circular microtubes used in this study are commercially available glass precision capillaries of Drummond Scientific make (catalog name: Wiretrol II) while rectangular geometry microchannels are of VitroCom Inc. make (catalog name: Microcells). Both plain glass substrates and microchannels are of Type II soda lime glass; hence the surface coating on both surfaces will be chemically same.

The below mentioned generalized procedure was followed to obtain the coatings of various surface wettability.

Table 4 lists the values of the parameters and chemical mixture composition used for coatings realized in this study. All the channels are pre-cleaned as per standard RCA cleaning procedure. The precursor solutions are prepared by mixing the chemicals and stirring vigorously for time t_1 . After adding specified amount of hydrolysis solution, the resultant solution is kept stirring for time t_2 to result in sol. The pretreated glass microchannels (and glass slide substrates) are dip coated in sols for n times, using a withdrawal speed of V approximately. After coating, the samples are dried at ambient temperature for 5 min and then heat treated at T_1 °C in air for time t_3 and at T_2 °C in air for time t_4 . The surfaces obtained were physically and chemically stable (in air and water) up to high temperatures till 500 °C.

Table 4- Chemical mixture composition and coating procedure parameters for various surface wettability sol-gel dip coatings obtained in this study

Wettability of Surface	Precursor chemical solutions	t_1 hr	Hydrolysis solution	t_2 hr	Withdraw speed V (mm/s)	n	T_1 °C	t_3 hr	T_2 °C	t_4 hr
Super-hydrophilic ⁵⁰	Aluminum tri- <i>sec</i> -butoxide, Iso-propyl alcohol, Ethyl acetoacetate	3	Water & Isopropyl alcohol	-	2	1	400*	1/6	500	1/6
Hydrophobic ⁵¹	Perfluoroalkylsilane, Tetraethoxy-silane, Ethanol	1/3	Distilled Water & HCl	2	0.3	4	120	1/3	250	1
Super-hydrophobic ⁵⁰	Aluminum tri- <i>sec</i> -butoxide, Iso-propyl alcohol, Ethyl acetoacetate	3	Water & Isopropyl alcohol	-	2	1	400*	1/6	400	1/6

* After heat treatment @ T_1 °C for time t_3 , coated glass microchannels were dipped in boiling water for 10 min, dried and subjected to further heat treatment @ T_2 °C for time t_4 .

4.1.3 Contact Angle Measurement System

Static contact angles were measured using sessile-drop method by AST VCA Optima XE surface analysis system. The system uses automated gray-scale imaging and performs a droplet base-line determination and curve-fit to calculate the contact angle. 2 μL of DI water is dispensed into needle from gas-tight 100 μL syringe (Make: Hamilton), using a syringe control command in Optima software. The glass substrate is raised slowly until it touches water droplet. Once glass substrate is in contact with water droplet, it is retraced slowly and image of water droplet on surface is captured after 5 sec. Mean contact angle of surface is computed using average of measurements.

Figure 4.1.3 (a) illustrates the contact angle values of water on sample coated surfaces with $\pm 2^\circ$ accuracy. To ascertain the stability of coated thin film surfaces on its continuous presence in water, contact angles of sample substrates are measured over a period of time and illustrated in Figure 4.1.3 (b). As observed from the figure, contact angle remained within measurement uncertainty of $\pm 2^\circ$ and hence it can be concluded that thin film coatings are stable in water.

Fig.4.1.3 (a) Measured values of Contact angle of various surface wettability surfaces. a) Super-hydrophilic (CA 20°) b) Superhydrophobic (CA >135°) c) Plain glass (CA 60°) and d) Hydrophobic (CA 105°)

Fig.4.1.3 (b) Plot of Static Contact angle of coated surfaces Vs No. of days (time) present continuously in air and water

4.2 Measurement of Microchannel Dimensions

Table 5 indicates the size of circular and square microchannels investigated in this study. The length of the microtubes is measured with digital calipers. As the square microchannels are commercially supplied precision channels, no measurements are carried out to confirm nominal cross sectional dimensions.

To measure the nominal diameters of the circular channels, image processing method is employed. Images of cross-section of circular tube along with known 700 μm and 1000 μm square channels are taken by high-spatial resolution camera fitted onto microscope. Care is taken to ensure the ends of the channels are in same plane by inserting microtubes parallel to each other.

Figure 4.2 shows images of circular channel taken alongside 1000 μm square channel. Using National instruments image processing software, area and length measurements are carried out to obtain the dimensions of the circular channel relative to square channel. To minimize errors, both 700 μm and 1000 μm square channels are used for the analysis. All the calculated measurements are found within +/- 5% range of the average.

Table 5- Specification of microchannels used in this study

Sl. No	Channel Geometry Description	Dia (in μm)	Hydraulic Diameter (μm)	Length (mm)	Make & Type
1	Circular	490	490	90	Drummond Scientific (Type II Soda Lime glass)
2		565	565	127	
3		690	690	90	
4		700	700	127	
5		925	925	127	
6		1090	1090	95.25	
7		1100	1100	90	
8	Square	400	400	102	Vitrocom Microcells (Type II Soda Lime glass)
9		700	700	102

Fig. 4.2. Snapshots of circular channels (of unknown diameter) taken along with square channels of known dimensions for image processing. Left: 700 μm circular channel with 1000 μm square channel. Right: 1100 μm circular channel with 1000 μm and 700 μm square channels

4.3 Experimental Setup

The experimental setup used in this study is designed for adiabatic co-current flow of air-water mixtures in either round or rectangular horizontal tubes. Test setup, shown below in Figure 4.3 (a), consists of water and dry air supplying lines, visualization system, differential pressure measurement setup and synchronized image & data acquisition system. The mass flow rates of the single phase (water and air) flows is calculated inversely using the laminar flow theory by measuring pressure drop across a microtube (ID 1095 μm , L 95.25 mm) mounted between an upstream supply sources and downstream two-phase flow test section. Photograph of experimental setup is shown in Figure 4.3 (b).

Fig. 4.3 (a) Experimental setup layout

Fig. 4.3 (b) Photograph of Experimental setup

A syringe pump infuses de-ionized distilled water at known flow rate in pump mode while dry air is supplied by regulating air cylinder supply. The syringe pump, capable of delivering high precision mass flow rates with high pressures, maintained pressures constant over each test run with dual stage air pressure regulator. Dry air from cylinder regulated to 50 psi is used as the supply. It is then controlled through a precision regulator (0 – 15 psi range) for maintaining test flow rates and filtered in the downstream before flow rate measuring microtube section.

Calibrated differential pressure transducers (Wet-Wet DPT, Make: Druck Ltd., Input: 9-30 VDC, Output: 4-20 mA DC) are used to measure pressure drops of both single phase flows and two-phase pressure drop. Details of the calibration are listed in Appendix A. The transducer output current varied linearly with applied pressure over its operating range (0 - 0.1 MPa). Uncertainty in pressure measurement is 0.25% of the maximum reading. The transducer is checked for its initial value of 4mA current output prior to running the test, and confirmed

following each test. Flow circuit including microchannel test section along with calibrated pressure transducers is shown in Figure 4.3(c).

Fig. 4.3 (c) Photograph of Experimental flow circuit

The experimental test microchannel sections are primarily of two different geometries (circular & rectangular) of different surface wettability conditions (superhydrophilic, plain – hydrophilic, hydrophobic and superhydrophobic). The two phases (water and air) meet at the entrance region and naturally generate converging-nozzle type entrance flow shown along with image in Figure 4.3 (d). Entrance losses at two-phase flow set-up are minimized because of the converging-nozzle type flow. In order to minimize the exit losses and its effects on pressure drop measurement, microchannel of similar surface wettability and geometry is included on the downstream side of the two-phase flow in test section. Thus entrance and exit effects on pressure drop are minimized significantly, by using this microchannel test section.

Fig. 4.3 (d) Sketch of two-phase flow test section along with entrance and exit sections. Inset: air-water flow mixing region

Two thermocouples are used to measure the temperature of the ambient near and away from the test section to verify isothermal conditions. Before starting experiment, pressure drop across all microchannels is maintained zero. Flow through the test section is initiated by starting syringe pump at a given constant flow rate Q .

The flow regime of the two-phase flow is confirmed with visualization of the flow patterns, recorded by scientific CCD camera of high resolution and shutter speed (Sony XCL-X700, 1024 X 768 pixels, 1/3-type progressive scanning, 10 bit depth, 1/20000 shutter speed, 30 fps) mounted on stereo microscope (SMZ-U 1000). The backdrop is illuminated by goose neck fiber optic guides connected to two independent 150W and 250W cold cathode quartz halogen fiber optic illuminators. Gooseneck guides are used to minimize heat input to the fluids from illumination. Temperature rise, due to illumination from gooseneck light sources, near the test section is observed to be within $\pm 2^\circ\text{C}$ for all the experimental runs. To record enlarged images of the two-phase flow inside the microchannel, a microscope objective lens (0.5X) is coupled to video camera or digital camera. Synchronized image and data acquisition is carried out by LabVIEW program.

4.3.1 Synchronized Data and Image Acquisition

The experimental data of pressure drop and temperature are acquired using data acquisition system, comprising National Instruments - SCXI 1000 DAQ chassis, SCXI 1102 C DAQ module, TBX-1303 auxiliary connector block, PCI 6052E, into personal computer using a LabVIEW program. As the TBX connector block does not support current measurements for data acquisition, current output from pressure transducer (mA DC) is acquired using Ohm's law by measuring voltage across $490\ \Omega$ precision resistor connected in series. This DAQ system offers 0.015% dcV accuracy of maximum reading at Gain 1 while sampling frequency can be varied in LabVIEW program. Images are acquired from Image acquisition setup comprising National Instruments camera link module frame grabber, PCIe 1429 into personal computer enabling synchronized data and image acquisition. LabVIEW program, enclosed in Appendix B, is written in parallel threads to enable synchronized data and image acquisition.

4.4 Experimental Conditions

All experimental investigations are carried out at room temperature. As shown in Table 1, the flow velocities of air and water supply correspond to laminar regime. Hence the current experimental investigation is laminar-laminar air-water two-phase flow in horizontal microchannels at adiabatic conditions.

Table 6- Experimental conditions

Parameter	Type	Condition
Gas	Dry Air	Laminar regime ($50 \leq Re_{air} \leq 250$)
Liquid	De-ionized water	Laminar regime ($5 \leq Re_{water} \leq 30$)
Temperature	-	Adiabatic (Room temperature)

4.5 Thin Liquid Film Thickness Measurement Setup

We have carried out experiments to measure the order of thickness of thin liquid film, present between gas bubble and solid channel wall, for various surface wettabilities. A known volume of water is infused into the circular microchannels (925 μm , 1090 μm and 1580 μm) using gas tight precision syringe followed by supply of humidified air at experimental flow rate conditions. The volume of water drained off the tube is estimated by measuring the Petri dish in the Microbalance (Make: A&D, Resolution: 0.01/0.1 mg, Range: 101/250 g) before and after liquid water drain off. Volume of liquid water retained in the microchannel is obtained by the difference between total volume of liquid water injected and volume of liquid drained. Assuming that liquid water retained in the microchannel remains as liquid film of uniform thickness along the channel length, thickness of the liquid film (δ) is calculated.

Fig. 4.5 Experimental setup for measuring thin fluid film thickness in microchannels

4.6 Methodology for Analyzing Experimental Data

In the present study we limited ourselves to characterization of pressure drops of two-phase flow in microchannels of various surface wettabilities and geometries. Methodology shown in Figure 4.6 (a) is employed in this study. Along with simultaneous single phase pressure measurements, two-phase flow pressure drop is measured. The flow regime of the two-phase flow is confirmed with the visualization of the flow patterns, recorded by the camera. Volumetric flow rates of single phase flows are calculated from pressure drop measurements using laminar flow theory for microchannels.

To carry out dynamic contact angle analysis besides flow regime confirmation, video files are processed using VirtualDub software for enhanced fluid-fluid-solid interface images. National Instruments DIAdem software is used for synchronized data and image analysis. As shown in Figure 4.6 (b), the video can be played frame by frame while a tab and pointer on the pressure drop plot displays the measure two-phase flow resistance.

Fig. 4.6 (a) Flow chart of methodology for experimental data and image analysis

Fig. 4.6 (b) Synchronized video and data analysis using National Instruments DIAdem software

CHAPTER 5

EXPERIMENTAL RESULTS AND DISCUSSION

In this section we will discuss the experimental results obtained for the following cases: two-phase flow resistance variation with water and air flow rates, dynamic contact angle variation with surface wettability, two-phase flow resistance variation with surface wettability, geometrical effects on two-phase flow resistance, two-phase flow resistance variation with slug frequency, and thin liquid film thickness variation with surface wettability.

5.1 Two-phase Flow Resistance Vs Single Phase Fluid Flow Rates

Experiments are conducted at various air and water flow rates. Figure 5.1 illustrates the flow resistance for two-phase slug flow in 700 μm diameter and 127 mm long uncoated plain circular glass capillary test section (of CA $\theta \sim 60^\circ$). The upper trace in Figure 5.1 is two-phase pressure drop while the middle and lower traces represent the corresponding volume flow rates of water and air respectively. All the striking features of pressure drop pattern mentioned by Son & Allen^{20,21}, briefly sudden disruption of flow resulting in multiple contact line motion followed by Taylor plug movement with a thin liquid layer on the wall, are observed. It can also be observed that the volumetric flow rate of the liquid phase generally varies inversely with two-phase pressure drop across the test section. Also there is a time lag in the response of volumetric flow rate with the

two-phase pressure drop for sudden changes in pressure. This time lag is associated with the compression and decompression of the gas phase between the liquid slugs.

Fig. 5.1 Variation of Two-phase flow pressure drop with air and water volumetric flow rates for flow in 700 μm plain circular channel (hydrophilic, $\theta \sim 60^\circ$). Upper trace: two-phase pressure drop, middle trace: water volumetric flow rate and lower trace: air volumetric flow rate

The moving contact lines dissipate much more energy than does the inertia and viscous dissipation of the liquid slug motion resulting tremendous increase in pressure drop. Also the pressure drop from surface energy for a slug train with N slugs can be correlated with modified Young's equation (14 a & b).

$$\begin{aligned}\Delta p_{capillary} &= \sum_{i=1}^{2N} \frac{2\sigma \cos \theta}{R} \\ \Delta p_{capillary} &= \sum_{i=1}^{2N} \left(\frac{\sigma \cos \theta_{adv}}{R} - \frac{\sigma \cos \theta_{rec}}{R} \right)\end{aligned}\tag{14 a) \& (14 b)}$$

5.2 Dynamic Contact Angle Vs Surface Wettability

Two-phase flows are distributed into several distinct flow patterns depending on gas and liquid flow rates. Only slug flow, which is commonly encountered in membrane electrode fuel cells, is studied. Transitions between regimes are predicted as a function of liquid and gas flow rates. For both circular and square channels, transitions between each regime occur for fixed values of the liquid volumetric fraction α_L defined as

$$\alpha_L = \frac{Q_L}{(Q_L + Q_G)}\tag{15}$$

Over the range of flow rates investigated (α_L : 0.007 to 0.03) slug flow is observed in all sizes of channels used in this study, independent of surface wettability and geometric configurations. This case is different from the case of macrochannels and minichannels where differences in the relative effects of gravitational, shear, and surface tension forces cause the transitions to be dependent on the channel diameter.²²

For liquid volumetric fraction α_L of 0.028, Figure 5.2 illustrates the contact line of the air-water two-phase flow observed in 700 μm circular channels of superhydrophilic, plain, hydrophobic and superhydrophobic wettabilities. For each wettability configuration, row **i** indicates the snapshot of the flow selected for processing. Images are post processed in VirtualDub software while dynamic contact angle measurements are carried out using National Instruments Vision software.

Superhydrophobic (Circular)

Circular channel (Φ 700 μ m, L 127 mm), Flow Conditions- $Q_G = 1.65 \times 10^{-6} \text{ m}^3/\text{s}$, $Q_L = 3.9 \times 10^{-8} \text{ m}^3/\text{s}$, i.e. $\alpha_L = 0.028$					
Superhydrophobic			Superhydrophilic		
θ_i	θ_d	$\theta_c - \theta_d$	$\theta_c - \theta_d$	θ_d	θ_i
		$150 - \theta_d$	$25 - \theta_d$		
173°	7°	143°	2°	23°	157°
133°	47°	103°	-15°	40°	140°

a) Dynamic contact angle analysis for superhydrophobic channel

Superhydrophilic (Circular)

b) Dynamic contact angle analysis for superhydrophilic channel

Hydrophobic (Circular)

Circular channel (Φ 700 μ m, L 127 mm), Flow Conditions- $Q_G = 1.65 \times 10^{-6} \text{ m}^3/\text{s}$, $Q_L = 3.9 \times 10^{-8} \text{ m}^3/\text{s}$, i.e. $\alpha_L = 0.028$					
Hydrophobic			Plain		
θ_i	θ_d	$\theta_c - \theta_d$	$\theta_c - \theta_d$	θ_d	θ_i
		$105 - \theta_d$	$60 - \theta_d$		
158°	22°	83°	35°	25°	155°
134°	46°	59°	14°	46°	134°

c) Dynamic contact angle analysis for hydrophobic channel

Plain (Circular)

d) Dynamic contact angle analysis for plain channel

Fig.5.2 Advancing and receding dynamic contact angles of slug flow observed in 700 μ m circular microchannel of various wettabilities at α_L of 0.028. For any given surface wettability, snapshot of flows are indicated in rows as mentioned, row i) Snapshot of flow, ii) interface and dynamic contact angle for advancing front, iii) interface and dynamic contact angle for receding front

Rows **ii** and **iii** for all wettability configurations in Figure 5.2 represent interface angles (θ_i), measured at the air-water interface through air phase, of the advancing and receding fronts of the slug flow respectively. Hence dynamic contact angle (θ_d) of the slug is $180-\theta_i$. The values of dynamic contact angle and its deviation from static contact angle are tabulated in a table in between the figures. Measurements for advancing interface and receding interface are highlighted in first and second rows of the table respectively.

For superhydrophilic and plain microchannels, advancing and receding interface of slug flow are similar in shape and the dynamic contact angle calculated through air phase is within the same range. As the surface energy decreases the advancing and receding interface of the slug flow vary in shape and thus the angles. For flows in microchannels with increasing hydrophobicity from SHPL to SHPB, dynamic contact angle deviation from its static value increases from -5° to 120° . Thus deformation of gas-liquid interface increases with hydrophobicity.

As was found by Qian et al.⁵² and Weiqing Ren et al.⁵³, Young's stress in the contact line region is dominant even in cases when the dynamic contact angle deviates only slightly from the static contact angle (denoted by $\theta_c-\theta_d$). As observed in the analysis, dynamic contact angle deviates significantly from static contact angle with surface wettability. Thus Young's stress becomes predominant in the contact line region for superhydrophobic surfaces. However in a dynamic situation, the stresses near the wall including the unbalanced Young's stress and the normal stress jump across the contact line region and acts as external forces on the contact line. From this discussion it can be concluded that contact line motion is an activated process at low capillary number (superhydrophobic surfaces), low driving force. The contact line motion becomes diffusive at large capillary number (superhydrophilic surfaces). Normal stress jump is important to account for the force balance inside the contact line region. Young's stress dominates over the viscous

stress inside the contact line region. Hence increase in Young's dynamic stress with surface wettability is primary cause for flow pattern variation and instabilities.

Evident from the Figure 5.2, difference in advancing and receding contact line is significant in hydrophobic and superhydrophobic cases. Through flow visualization of two-phase flow in superhydrophobic microchannels, it can be inferred that slip occurs at the coated surfaces as gap is observed near the surface. The results confirm Meinhart⁵⁴ observations that slip occurs over a gap near the surface rather than directly on the solid surface and the gap forms when a hydrophobic liquid flows over a hydrophilic surface and vice versa. Further flow resistance due to instabilities and break up of fluids will be significantly higher.

5.3 Two-phase Flow Resistance Vs Surface Wettability

Flow resistance caused by frictional force in two-phase flow is a parameter of prime interest in this study. Figure 5.3.(a) below indicates the variation of averaged two-phase flow resistance against static contact angle for two-phase flow in 700 μ m microchannels of both square and circular geometries, for various liquid volumetric flow ratios. Shape of markers is used to represent geometry of the channel. As evident, for same value of α_L (accurate with variance of +/- 3%) and wettability, two-phase flow resistance in circular channels is higher than that in square channels. Further from flow visualization, it is observed that slug flow with Taylor bubbles is observed for low contact angle while the flow tends to instability for higher contact angles.

Figure 5.3.(b) is another graphical representation of above results. The upper row indicates the two-phase flow resistance for circular channels while bottom row for square channels. Different surface wettabilities are highlighted by different colors and are shown along each column. Two-phase flow resistance is plotted against α_L .

As evident, for a given value of α_L and wettability, two-phase flow resistance in circular channels is higher when compared to square channels while the flow tends towards instability in for both geometric superhydrophobic channels. Instability of the flow observed in hydrophobic circular channels is relatively higher than that observed in hydrophobic rectangular channels. Further from flow visualization, slug flow with Taylor bubbles is observed in superhydrophilic channels.

Fig. 5.3 (a) Plot of two-phase flow resistance per unit length Vs surface wettability, for flows at different liquid volumetric flow rates α_L . (700 μ m circular microchannel, L=127mm).

Fig. 5.3 (b) Plot of two-phase flow resistance Vs α_L in 700 μ m microchannel at various α_L and surface wettabilities (Circular channel length = 127 mm, Square channel length = 102 mm)

The results of flow pattern variation with surface wettability are highlighted in Table 7. While smooth slug flow with shorter bubble length is observed for flows in superhydrophilic channels,

the flow tended more towards instable transition flow for superhydrophobic channels. At higher α_L conditions in hydrophobic and superhydrophobic channels, intermittent appearance of spherical droplets is observed as regular phenomena. Completely snapped off slug appearance, denoted by slug frequency f_s , decreased as hydrophobicity of microchannels increased; however instability of the flow increased significantly.

Table 7- Characteristics of flow pattern of Two-phase flow in 700 μ m circular microchannel for value of α_L of 0.028

a) Superhydrophilic	Slug Flow with shorter bubble length; $f_s = 12$
b) Plain (hydrophilic)	Slug Flow with longer bubble length; $f_s = 10$
c) Hydrophobic	Liquid ring flow with intermittent appearance of spherical droplets; $f_s = 9$
d) Superhydrophobic	Highly instable transition flow with frequent spherical droplets (shown in the inset); $f_s = 7$

For super-hydrophilic and plain microchannels, advancing and receding interface of slug flow are similar in shape and the dynamic contact angle remains almost in the same range for advancing and receding interfaces. As the surface wettability decreases (i.e. as contact angle increases), the advancing and receding interface of the slug flow vary in shape. From the Figure 5.2, increased deformation of interface leads to higher drag force along with higher shear forces between the interfaces. As evident from the figure, difference in advancing and receding contact line of slug is significant in hydrophobic and superhydrophobic cases. As hydrophobicity increases, difference in dynamic contact angle at advancing and receding interface is higher (say $\Delta\theta_d = \theta_{d\text{adv}} - \theta_{d\text{rec}}$). Value of $\Delta\theta_d$ varies from 30° to 10° for superhydrophobic surfaces to superhydrophilic surfaces. Hence,

it can be concluded that increased variation of dynamic contact angle from its static value at advancing & receding interfaces, leads to increase in two-phase flow resistance.

Using Young's modified equation (14 b); the contribution of capillary force for each air-water slug towards two-phase flow resistance is calculated with surface wettability and tabulated in Table 8. Hence as $\Delta\theta_d$ increases, stress experienced by slug on advancing and receding fronts increases significantly which contributes for increase in two-flow resistance in superhydrophobic channels. Hence the contribution of dynamic contact angle to two-phase flow resistance increases with hydrophobicity.

Table 8 – Flow resistance of individual slug in various surface wettabilities microchannels for flow of α_L of 0.028 in 700 μ m circular channel

Description	Advancing dynamic contact angle ($\theta_{d\text{ adv}}$)	Receding dynamic contact angle ($\theta_{d\text{ rec}}$)	Diff = $\text{Cos}(\theta_{d\text{ adv}}) - \text{Cos}(\theta_{d\text{ rec}})$	Dynamic flow resistance for individual slug in Pa ($\sigma^*\text{Diff}/R$)
Superhydrophilic	40	22	-0.161	33.48
Plain	45	25	-0.199	41.39
Hydrophobic	47	24	-0.231	48.12
Superhydrophobic	47	7	-0.310	64.53

5.4 Two-phase Flow Resistance Vs Geometry

Figure 5.4 represents variation of two-phase flow resistance with surface wettability and geometries for similar flow rate conditions of α_L of 0.028 in 700 μ m hydraulic diameter channels. It can be inferred from above Fig. 5.3.(a) and 5.4, that geometry of the channel has significant influence two-phase flow resistance. On synchronized analysis of flow visualization data with pressure drop, elucidated the mechanism geometry influenced the flow resistance. Geometry influenced slug frequency. For similar flow conditions of α_L and wettability, as the geometry of microchannel changes from circular to square, completely snapped off slug frequency decreases while instabilities remained on the same order.

Fig.5.4 Two-phase flow resistance in 700 μ m microchannels of various surface wettabilities (static contact angles) and geometries at liquid volumetric flow rate α_L of 0.028

5.5 Influence of Slug Frequency on Two-phase Flow Resistance

As reported by Kawaji²⁷⁻²⁹, we have not observed bubbly flow regimes for two-phase flows established over the flow rate ratios α_L of 0.007 to 0.03. Figure 5.4 represents the flow regimes observed in 700 μ m circular channel at α_L of 0.028. Slug flow regime is observed in superhydrophilic channels while flow tends towards transition for superhydrophobic channels.

Total number of slugs present in the microchannel test section at given instant of time is calculated assuming uniform slug velocity. Ratulowski and Chang⁵⁵ proposed an expression for the pressure drop, assuming that the pressure drop is the sum of the pressure across bubbles and

liquid slugs. They found an expression for the pressure that is proportional to the number of bubbles in the channel nb .

As observed in the figure, the slug frequency increases with increase of liquid volumetric flow rate and contributes towards increased flow resistance. The slug frequency observed for flows in superhydrophilic channels is higher than that observed in superhydrophobic channels. However flow resistance increases with hydrophobicity. Though instabilities contribute to the increase in flow resistance, significant increase in flow resistance can be accounted by the liquid film thickness present between the air and the microchannel wall. As no-slip and apparent slip has been noticed by Meinhardt⁵⁴ for hydrophilic and hydrophobic channels, it can be ascertained that there exists a thin liquid film for hydrophilic channels whose thickness decreases with increase of hydrophobicity. For the flow rates considered in this study, Taylor bubble flow is observed in superhydrophilic channel which flows smoothly on a thin fluid film present on channel wall. Hence no three-phase (air-water-solid wall) interface is observed at air-water contact line for superhydrophilic surfaces. However for super-hydrophobic surfaces, very thin fluid film facilitates three-phase interface at the air-water contact line. It can be concluded that thin liquid film is an important parameter besides slug frequency for accounting variation of two-phase flow resistance with surface wettability.

5.6 Thin Liquid Film Vs Surface Wettability

In Slug flow, the bubble length is far larger than the channel height ($d \gg h$). Bubbles are surrounded by liquid so they do not touch the channel walls. For long bubbles the film between gas and solid may dry out. Further many studies have inferred variation of thickness of liquid film with surface wettability.

Experiments are carried out to understand the order of thickness of thin liquid film for various surface wettabilities configuration. A known mass (or volume) of water is injected into the microchannel kept in Petri-dish in near horizontal position using precision syringe. While water remains in the channel, free end of the channel is connected to saturated air supply. As air at the end of microchannel test section in actual experiment conditions is completely saturated, similar condition is established by supplying humidified air at main two-phase experimental flow rate conditions.

Fig.5.6 Variation of thickness of liquid film with surface wettability for various circular channels

The water which drained off the other end of the tube is collected into a separate Petri-dish. Mass of drained water is calculated using a precision micro-balance. Applying mass conservation, the mass of liquid retained in the microchannel is estimated by deducting drained off liquid mass from the total mass. Assuming that water remains as a thin liquid film of uniform thickness along the inner surface of the channel length, thickness of the liquid film (δ) is calculated. Figure 5.6

represents variation of thickness of thin liquid film with surface wettability for various microchannels. Thickness of fluid film in superhydrophilic channels is an order of magnitude higher when compared to that in superhydrophobic channels. Hence thickness of liquid film is an important parameter which explains and accounts for two-phase flow resistance with surface wettability.

5.7 Conclusion

In this present work we aimed to provide data for understanding the role of surface wettability on the two-phase flow characteristics in microchannels. Two-phase slug flow in microchannels is experimentally characterized using flow visualization and two-phase pressure drop measurement. Flow visualization is used extensively to ascertain the influence of surface wettability on dynamic angle and thus flow resistance. Also geometry of the channel affects snapped off slug frequency of two-phase flow for similar flow rate conditions. Liquid film thickness variation with surface wettability, which is determined experimentally, influences the flow characteristics and resistance. Effects of wettability and geometrical variations on two-phase pressure drop are dramatic. Hence, characterization of flow resistance of two-phase flows in microchannels has to be carried out based on the appropriate parameters like surface tension, liquid film thickness, and dynamic contact angle besides thermo-physical quantities.

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APPENDIX A

Pressure transducers used in this study are calibrated using in-house set up available at Gas Dynamics and Propulsion Lab, University of Cincinnati. Figure 1 represents calibration curves for the pressure transducers used to measure air, water and two-phase flow pressure drops. Using curve fitting method for obtained output values, correlation curves are obtained.

Fig.1. Calibration curve plots of pressure transducers used in this study.

APPENDIX B

In this section, we include the screenshot of the LabVIEW program written for synchronized data and image acquisition in this experimental study. (National Instruments LabVIEW 8.0 platform is used).

Fig.1. Screenshot of Front panel of Data & Image Acquisition program: Acquire Mode

Fig.2. Screenshot of Front panel of Data & Image Acquisition program: Save Mode

Fig.3. Screenshot of block diagram of Data & Image Acquisition program: Acquire Mode

Fig.4. Screenshot of Front panel of Data & Image Acquisition program: Acquire Mode