Effects of solid-liquid intermolecular interactions on liquid flow and heat transfer in microtubes

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Abstract. This paper investigates the laminar flow and heat transfer characteristics of liquid in microtubes by considering the solid-liquid intermolecular interactions. We propose an apparent viscosity model of liquid from the wetting theory and the Hamaker theory to account for the interface effect. The numerical simulation considering this model is conducted to study the flow and heat transfer characteristics of liquid in microtubes. Simulation results indicate that the flow and heat transfer characteristics such as velocity distribution, pressure drop, flow friction and heat transfer coefficient deviate from the predictions of the conventional theory. The deviation increases as the hydrophilicity or hydrophobicity increases, and decreases as the diameter of the microtube increases.

1. Introduction

Microscale liquid flow and heat transfer have attracted much attention due to the rapid development of Micro-Electro-Mechanical System (MEMS) and micro-fluidic devices. A deep understanding of microscale flow and heat transfer characteristics is also critical for the design and the process control of MEMS and micro-fluidic devices. It has been found that the flow and heat transfer characteristics in microchannels often deviate significantly from the predictions of conventional theory. Peng et al. [1,2] found that the flow friction behaviors and the Nusselt numbers were depart from the classical correlations. The geometrical parameters, such as hydraulic diameter and aspect ratio, were found to have important effects on the flow behavior. Mala and Li [3] observed that the flow behaviors depend on the material of the microtubes, the pressure drop in stainless steel tubes is lower than that in fused silica tubes under the same flow rate. The pressure gradient is higher than that predicted by the conventional theory, and the deviation increases as the diameter of the microtubes decreases. Qu et al. [4] proposed that the measured higher pressure gradient and flow friction maybe due to the effect of surface roughness of the microchannels. They proposed a roughness viscosity model to interpret the experimental data. Xu et al. [5] found that flow characteristics in microtubes with diameters of 16 μm and larger ones are in agreement with the classical theory. However, as the diameters decreased to 5 and 2 μm, the nonlinear flow characteristics prevail and the results indicate significant departure of flow characteristics from the predictions of the conventional theory, and the smaller the diameters, the larger the departure. An experimental study by Wang et al. [6] also indicated that the flow characteristics of deionized water in microtubes with diameter of 14.9 μm are in agreement with the classical theory. But for hydrophilic and hydrophobic microtubes with smaller diameters, the results show significant departure from the prediction of conventional theory. The discrepancies are influenced by wettability of the microtubes and increase as the Reynolds number decreases.
It can be concluded that the flow and heat transfer characteristics of liquid in microchannels are affected by geometrical parameters, surface roughness, and properties of the liquid and the solid wall. Therefore, the solid-liquid interfacial effects may play an important role in the microscale flow. For macroscale flow, the interfacial effects are ignored usually due to the scope of interfacial effects is far less than the hydraulic radius of the channel. However, when the channel size down to microscale these effects cannot be ignored.

Mala et al. [7,8] studied the effects of interfacial electric double layer (EDL) caused by the electrostatic interaction on liquid flows through a microchannel. The EDL act against the liquid flow resulting in a higher apparent viscosity. The apparent viscosity can be several times higher than the bulk viscosity of the liquid. The EDL results in a reduced velocity of flow than in conventional theory. Experimental indicate that pressure gradient and friction factor were found to be significantly higher than that without electro-viscous effect caused by EDL [9,10].

Similar to electrostatic interactions, the solid-liquid intermolecular interaction, i.e., Vander Waals force also affects the liquid apparent viscosity near the wall. It had been found that the viscosity of liquids in thin films between two surfaces can be much different from those in bulk region, the viscosity of liquids decrease from solid surface to bulk continuously [11,12]. Lv et al. [13] found that the liquid viscosity in microcrevices increases as the thickness of microcrevice decreases. They also found that the liquid viscosity in small microcrevice is affected by the solid surface and the liquid molecule structure. The molecular dynamics simulation results also indicated that the liquid viscosity can be inhomogeneous near the wall [14-16]. Because the magnitude of the surface wettability is also determined by the strength of the intermolecular interactions, surface wettability may plays a role in the microscale flow.

Under the above motivation, in this work, we theoretically investigate the effects of solid-liquid intermolecular interactions on liquid laminar flow and heat transfer in microtubes. We propose a model of the liquid apparent viscosity in the region near the interface to account for the interface effect. Based on the simulation results, we attempt to explain the effects of solid-liquid intermolecular interactions on flow and heat transfer characteristics.

2. Model of liquid apparent viscosity
The value of liquid viscosity is related to the strength of the intermolecular interactions. In the region near the solid-liquid interface, the intermolecular interactions acting on the liquid molecules include not only liquid-liquid intermolecular interactions but also solid-liquid intermolecular interactions, so the viscosity of liquid adjacent to the interface is different from the viscosity in the bulk region.

As being shown in Figure 1, \( F_{LSA} \), \( F_{LLA} \), and \( F_{LLA} \) defined as the force acting on molecule A by the solid molecules, the upper liquid molecules and the nether liquid molecules respectively, and we have \( F_{LSA} = 0 \), \( F_{LLA} = F_{LLA} \). So \( F_A \), defined as the resultant force acting on molecule A, is equal to 0. Molecule B locates in the interface region, we can find \( F_{LSB} > 0 \), \( F_{LLB} > F_{LLB} \). If \( F_{LSB} + F_{LLB} > F_{LLB} \), then \( F_B > 0 \), the direction of \( F_B \) towards solid; if \( F_{LSB} + F_{LLB} < F_{LLB} \), then \( F_B < 0 \), the direction of \( F_B \) towards liquid. Molecule C locates on the interface, \( F_{LLC} = 0 \) and the value of \( F_L \) reaches the maximum value, the absolute value of the resultant force reaches the maximum. Therefore, the absolute value of the resultant force decreases as the distance away from the interface increases.
Figure 1. Force Analysis of molecules near the interface.

Assuming that the value of liquid viscosity is proportional to the strength of intermolecular interactions, the viscosity of liquid near the interface consists of two parts, one is the viscosity generated by the liquid-liquid intermolecular interactions, which is just same as the viscosity in the bulk region, and the other is the additional viscosity generated by the solid-liquid intermolecular interactions. In order to investigate the variation of viscosity, an apparent viscosity $\mu_a$ is defined to distinguish from the dynamic viscosity $\mu_0$, the apparent viscosity near the interface will be

$$\mu_a = \mu_0 + \mu'$$ (1)

Where, $\mu'$ is the additional viscosity.

On the interface, due to the value of viscosity is proportional to intermolecular forces, $\mu'$ can be written as $m\mu_0$, the value of $m$ can be calculated by

$$m = \frac{F_{LS} - F_{LL}}{F_{LL}}$$ (2)

As is well known, the strength of solid-liquid interactions can usually be gauged from the static contact angle: small angles indicate strong interactions, while large angles indicate weak interactions. We use the interface layer model of physical interface [17] to describe the static contact angle, according to this model, the liquid-liquid interaction force $F_{LL}$ is just the same as the internal surface force of liquid phase $\sigma_L^0$, and the solid-liquid interaction force $F_{LS}$ is just the same as the external surface force of liquid phase $\sigma_L^0$. The internal surface force of each phase can be considered as the surface tension of each phase. For liquid phase, we have

$$\sigma_L^0 = \sigma_L$$ (3)

Where, $\sigma_L$ is the surface tension of the liquid.

The interfacial tension defined by this model is the respective interfacial tension of two phases. The interfacial tension of the liquid phase $\sigma_{LS}$ is given by

$$\sigma_{LS} = \sigma_L^0 - \sigma_{LS}^0$$ (4)

From equation (3) and (4), we can obtain

$$m = \frac{\sigma_{LS}}{\sigma_L}$$ (5)

Unlike Young equation, the interface layer model of physical interface considers that the liquid and solid phase should be discussed independently when discussing wettability. Figure 2 shows the contact
angle and the liquid phase surface force system defined by the interface layer model of physical interface. The wetting equation of the liquid phase is

\[ \sigma_L \cos \theta + \sigma_{LS} = 0 \]  

(6)

Figure 2. Contact angle and tension in the interface layer model of physical interface. (a) wetting; (b) no wetting.

Substituting equation (6) into equation (5), we obtain

\[ m = \cos \theta \]  

(7)

Thus the value of \( m \) can be gauged from the static contact angle, small angles indicate strong interactions and large additional viscosity, while large angles indicate weak interactions and small additional viscosity.

Because the solid-liquid interactions decrease as the distance apart from the interface increases, the apparent viscosity in the interface region will be

\[ \mu_a(\delta) = \mu_0 + m\mu_0 L^n \]  

(8)

Where, \( m\mu_0 L^n \) is the additional viscosity; \( m \), the coefficient related to the properties of the solid surface and the liquid, increases as the solid-liquid interactions increase; \( L = l/\delta \), non-dimensional distance, where \( l \) is the distance away from the solid-liquid interface, \( \delta \) is the molecular separation; \( n \) is a coefficient.

According to Hamaker theory, the interaction free energy between two objects is related to the shape of the two objects. For example, the interaction free energy between a solid plate and a cube-type liquid micelles is proportional to \( l^2 \), the interaction free energy between a solid plate and a spherical liquid micelles is proportional to \( l^{-1} \), so we can get \( n \leq 3 \).

For a circular microtube, equation (8) can be rewritten as

\[ \mu_a(\delta) = \mu_0 + m\mu_0 \left( \frac{R - r}{\delta} \right)^n \]  

(9)

Where, \( R \) is the radius of the microtube; \( r \) is the distance from the axis.

3. Simulation results and discussion

In this work, water flowing in microtubes driven by an external force with a fixed inlet velocity is simulated by using FLUENT. In the simulation, the viscosity of water is determined by equation (9). The radius of the microtubes range from 0.1 to 1 μm.

According to equation (9), if \( m = 0 \), \( \mu_a = \mu_0 \), the Navier-Stokes (N-S) equations will be simplified to the conventional theory, so we use the simulated result of \( m = 0 \) to represent the conventional theory. All the data discussed in this section was taken from the fully developed region.
3.1. Apparent viscosity
As can be seen from Figure 3, the apparent viscosity of water is not a constant due to the solid-liquid interactions. If the solid surface is hydrophilic, i.e. \( m > 0 \), the apparent viscosity \( \mu_a \) of water near the wall is greater than the viscosity in bulk region \( \mu_0 \); if the solid surface is hydrophobic, i.e. \( m < 0 \), we can find that \( \mu_a < \mu_0 \) in the region near the wall. The value of \( m \) determines the maximum deviation between the apparent viscosity and the viscosity in bulk region. The values of \( m \) and \( n \) affect the effective range of influence on apparent viscosity, the larger the value of \( n \) or the smaller the value of \( |m| \), the smaller the effective range of influence. For strong hydrophilic material surface, the range of influence on the viscosity can up to tens or hundreds of nanometers.

3.2. Velocity distribution
As can be seen from Figure 4, without considering the effects of solid-liquid interactions, i.e. \( m = 0 \), the velocity distribution is just the same as the conventional theory and has \( u/u_m = 2 \) when \( r = 0 \). When \( m > 0 \), the apparent viscosity causes the velocity gradient near the wall to become smaller, so \( u/u_m \) is smaller in the region near the wall and is higher in the region near the microtube axis than the value calculated by the conventional theory. When \( m < 0 \), the deviation is opposite to that of \( m > 0 \). This deviation increases as the strength of hydrophilicity or hydrophobicity increases.

3.3. Pressure drop
As shown in Figure 5, the curves of pressure drop vs. mean velocity are linear as expected, when \( m > 0 \), the pressure drop is higher than that calculated by the conventional theory, the larger the value of \( m \) or the smaller the value of \( n \), the higher the pressure drop. From another perspective, for a fixed pressure drop, if the wall of channel is hydrophilic, the volume flow rate will be smaller than the conventional theoretical value; if the wall of channel is hydrophobic, the volume flow rate will be higher than the conventional theoretical value. Therefore, to obtain larger flow rate, the wall need to be treated to hydrophobic surface.

The pressure drop ratio is plotted in Figure 6 as a function of tube radius, where \( \Delta P_0 \) is the values of the case \( m = 0 \). We can see from Figure 6 that for tubes with a radius of hundreds of nanometers, the results show significant departure from the prediction of conventional theory, but the pressure drop ratio decreases as the pipe radius increases, the value of the ratio will be reduced to 1 when the radius is large enough, so for large diameter tubes the conventional theory is applicable. As can be seen from Figure 6,
the smaller the value of $n$ or the larger the value of $m$, the larger the radius of the microtube needed when the ratio reduce to 1.

Figure 5. Pressure drop vs. mean velocity for various $m$ and $n$ at $R = 0.1 \mu m$.

Figure 6. Pressure drop ratio $\Delta P/\Delta P_o$ vs. radius for various $n$.

3.4. Flow Friction
The flow behavior can be further interpreted in terms of some dimensionless parameters, such as Reynolds number $Re$, friction factor $f$ and friction factor constant $C$, which are defined as

$$Re = 2R \rho u_m / \mu$$  \hspace{1cm} (10)

$$f = 8\tau_w / (\rho u_m^2) = 4R\Delta P / (L\rho u_m^2)$$  \hspace{1cm} (11)

$$C = f Re$$  \hspace{1cm} (12)

Where, $u_m$ is the mean velocity, $\tau_w$ is the wall shear stress, $L$ is the length of the microtube.

Figure 7. Friction factor constant ratio vs. $m$ for various $n$ at $R = 0.1 \mu m$.

Figure 8. Friction factor constant ratio vs. radius for various $n$. 
In the conventional theory, the friction factor constant \( C \) is a constant value of 64, i.e. the ratio of \( C/C_0 \) is always equal to 1, but as can be seen from Figure 7, the value of the ratio is smaller than 1 when \( m < 0 \) and larger than 1 when \( m > 0 \), the ratio increases as \( m \) increases, i.e., the flow friction increases as the strength of the hydrophilic increases, and this trend increases as the value of \( n \) decreases. Friction factor constant ratios are plotted in Figure 8 as a function of radius. As can be seen from Figure 8, if the wall of the microtubes is hydrophilic surface, the value of the ratio is always larger than 1 and decreases as the radius increases, when the radius is large enough the value of the ratio will reduce to 1.

3.5. Heat transfer
Due to the viscosity of water is also related to temperature, the empirical correlation between viscosity of water and temperature should be considered in the calculation of \( \mu_0 \).

For a circular microtube, the Nusselt number \( \text{Nu} \) is defined as

\[
\text{Nu} = \frac{2Rq_w}{k(T_w - T_i)}
\]

(13)

Where, \( q_w \) is the wall heat flux, \( k \) is the thermal conductivity, \( T_w \) is the wall temperature, \( T_i \) is the mean temperature of the fluid and defined as

\[
T_i = T_w + \int_0^R u(T - T_w) r dr/\int_0^R ur dr
\]

(14)

The convective heat transfer process is closely related to the flow field, the change of the temperature gradient near the wall is similar to the change of the velocity gradient. Then, the change of the temperature gradient will affect the convective heat transfer coefficient and the Nusselt number. For a prescribed surface heat flux \( q_w \), as can be seen from Figure 9, the Nusselt number of the case of \( m > 0 \) is smaller than the conventional theoretical value, and decreases as the value of \( m \) increases, i.e., the heat transfer capacity of microtubes with hydrophilic surface is weakened. This trend increases as the value of \( n \) decreases.

As can be seen from Figure 10, the Nusselt number increases as the radius increases, but this trend decreases as the radius increases, when the radius is large enough we can infer that the Nusselt number will be a constant.

Figure 9. Nusselt number vs. \( m \) for various \( n \) at \( q_w=30000 \text{ W/m}^2 \), \( u_m=0.05 \text{ m/s} \).

Figure 10. Nusselt number vs. radius for various \( n \) at \( q_w=30000 \text{ W/m}^2 \), \( u_m=0.05 \text{ m/s} \).
4. Conclusions
In this paper, the effects of solid-liquid intermolecular interactions on liquid laminar flow and heat transfer in circular microtubes are studied. Our studies can be summarized as follows:

(1) In the region near the interface, the apparent viscosity is different from the viscosity in the bulk region due to the solid-liquid interactions. A model of the liquid apparent viscosity is derived from the wetting theory and the Hamaker theory, the value of apparent viscosity is related to the wettability of the solid surface.

(2) Simulation results indicate that flow and heat transfer characteristics in microtubes deviate from the conventional theory. For the hydrophilic surface, the apparent viscosity near the interface is greater than the bulk value, which results in a smaller velocity gradient near the wall. At a fixed flow rate, the pressure drop and the flow friction are higher than that predicted by the conventional theory, while the Nusselt number is less than that predicted by the conventional theory. For the hydrophobic surface, the trend is just the opposite from that of the hydrophilic surface. The deviation increases as the strength of hydrophilicity or hydrophobicity increases, and increases as the diameter of the microtube decreases.

(3) For microtubes, the hydrophilic surface will cause higher flow friction and weaker heat transfer efficiency than the hydrophobic surface. To enhance flow and heat transfer efficiency, the wall surface of the microchannels should be treated to a hydrophobic surface.

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References
[1] Peng X F, Peterson G P and Wang B X 1994 Exp. Heat Transfer 7 249-64
[2] Peng X F, Peterson G P and Wang B X 1994 Exp. Heat Transfer 7 265-83
[3] Mala G M and Li D Q 1999 Int. J. Heat Fluid Fl. 20 142-48
[4] Xu S L, Gao W L and Li D Q 2000 Int. J. Heat Mass Tran. 43 353-64
[5] Xu S L, Yue X A and Hou J R 2007 Chin. Sci. Bull. 52 849-54
[6] Wang F, Yue X A, Xu S L, Zhang L J, Zhao R B and Hou J R 2009 Chin. Sci. Bull. 54 2256-62
[7] Mala G M, Li D Q and Dale J D 1997 Int. J. Heat Mass Tran. 40 3079-88
[8] Mala G M, Li D Q, Werner C, Jacobasch H J and Ning Y B 1997 Int. J. Heat Fluid Fl. 18 489-96
[9] Ren L Q, Qu W L and Li D Q 2001 Int. J. Heat Mass Tran. 44 3125-34
[10] L Zhigang, G Ning, Z Chengwu and Z Xiaobao 2009 Exp. Heat Transfer 22 178-97
[11] Israelachvili J N 1986 J. Colloid Interf. Sci. 110 263-71
[12] Gee M L, McGuigan P M, Israelachvili J N and Homola A M 1990 J. Chem. Phys. 93 1895-906
[13] Lv P, Yang Z H, Hua Z, Li M Y, Lin M Q and Dong Z X 2016 Colloid. Surface. A 504 287-97
[14] Thomas J A and McGaughhey A J 2008 Nano. Lett. 82 788-93
[15] Suk M E and Aluru N R 2013 Rsc. Adv. 3 9365-72
[16] Ghobranian J and Beskok A 2016 Microfluid. Nanofluid. 20 121
[17] Zhang F T 2001 J. Colloid Interf. Sci. 244 271-81