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Effects of Real Surface Roughness on Fluid Flow in Nanochannels

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ABSTRACT
In the present study, a computer program based on a molecular dynamics scheme has been developed for simulating fluid flow in nano- and micro- channels with roughness. According to the previous studies of nanochannels flows, surface roughness has a great effect on the rheology of the flow. Therefore a more realistic surface roughness has been developed and its influence on the fluid flow has been investigated using Molecular Dynamics (MD) Simulation.

KEYWORDS: *Molecular Dynamic Simulation, Surface roughness, Interface wettability, Nanochannel*

INTRODUCTION
Studies of flow at nanosclae have attracted considerable attention with many important applications in nanofluidic devices, material science and biological systems. Compared with flow at conventional conditions, fluid flows at nanoscales have remarkable differences in hydrodynamic characteristics. The nanoscale flows are characterized by large surface-to-volume ratios, so that the molecular interactions at fluid–solid interface become particularly important. For macroscopic flows, the no-slip or stick boundary condition has been well established in the continuum regime. However, as the length scale over which the fluid velocity changes approaches the slip length, the fluids can undergo slip at the wall–fluid interface and the standard assumptions of the classical continuum theory with a no-slip boundary condition can break down.

The so-called slip boundary can be classified in various respects, e.g., fluids truly sliding over solid surfaces, apparent slip stemming from surface inhomogeneities, and complex interfacial effects with additional physics, etc. At various wall and fluid conditions, the behaviors of slip, no-slip, and multilayer locking have been observed [1].

Molecular dynamic (MD) method, a viable approach for investigation of the flow physics in nanoscale flows, distinguishes itself from other simulation methods by providing atomistic level direct numerical experiments that enable simulations with various physical conditions. Therefore, MD method can potentially address issues such as solid-fluid interfaces and interactions arising in the nanoscale regime. From a theoretical point of view, the parameters controlling the degree of slip are still largely unknown. Previous experimental studies have shown that the boundary slip is highly affected by the solid-fluid interaction energy or the wetting surface and the surface roughness [2,3,4,5,6,7,8,9]. Other numerical simulations also indicate that the boundary slip on the solid-fluid interface is mainly a function of the wall-fluid interaction strength, fluid/wall density and shear rate. [10 ,11]. Molecular Dynamics simulation of pressure driven flow in a nanochannel by Nagayama and Cheng revealed that the fluid behavior at the solid–liquid interface depends on the interface wettability and the magnitude of the driving force. [12]

The boundary conditions considered in most literatures are regarded as smooth surface. In practice, few surfaces are smooth for actual nanofluidic systems. As long as the characteristic length of the surface roughness is in the order of molecular structure, the effects of surface roughness on the rheology and fluid slip become important and a significant interfacial phenomena factor [13]. A rough wall can be intentionally fabricated with elements mounted on or grooves indented in solid surfaces. A few previous studies dealt with the effects of surface obstacles or grooves in nanoscale flows, e.g., Couette flow [14], electro-osmotic flow [15,16] and Poiseuille flow in which the flow behavior and slip length were examined [17]. In these cases, the roughness elements considered were of several σ (molecular length scale) in height and width. Both

roughness and cavitations are fundamental elements on a rough channel surface. In macroscopic flow, no divisions have been made between them. However, in micro channel flow, some difference has been reported on the flow friction between roughness and cavitations [18,19]. This rarely studied case was investigated in electro-osmotic flow [20] and Lennard-Jones fluid Poiseuille flow [21].

In all previous studies, roughness of the wall was modeled with periodic organized nanostrips of obstacle or grooves, while in the real world, the roughness of the nanochannel walls is comprised of random obstacle and grooves in the wall, rather than sequential well-arranged nanostrips which is mainly because of the methods used in fabricating nanochannel. Regardless of the different techniques employed in making the nanochannel, they all bear random surface roughness with different roughness amplitude. The summary of various surface roughness can be found in [22,23].

The purpose of this paper is to examine the effects of different wall factors on the fluid flow in a nanochannel by MD simulation. In this study two different rough surface were modeled, namely the organized and disorganized roughness. In addition, the flow is allowed to slip on the solid surface with altering the amount of wall/fluid interaction and the effect of surface roughness is investigated.

SIMULATION METHOD

Molecular dynamics simulation consists of the numerical, step by step, solution of the classical equations of motion, which for a simple atomic system may be written as:

$$m_i \ddot{\mathbf{r}}_i = \mathbf{f}_i \qquad \mathbf{f}_i = -\frac{\partial}{\partial \mathbf{r}_i} V \tag{1}$$

Where $\ddot{\mathbf{r}}_i$ is the acceleration of particle i, and V is of the form $V=V(r^N)$ where $r^N=(r^1,r^2,r^3,...,r^N)$ represents the complete set of 3N atomic coordinates. There is an extensive literature to determine the governing intermolecular potential experimentally or model theoretically. The most famous pairwise potential widely used is Lennard-Jones 6-12:

$$v^{LJ}(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \tag{2}$$

Where σ is molecular diameter and ε is the well depth of potential function.

The simulation is performed in a 3D channel undergoing poiseuille flow. The fluid is confined to flow between two solid planar walls parallel to xy plane. The upper solid surface is smooth while the bottom wall is decorated with various roughness atoms to account for simulation of different wall roughnesses. Figure 1 shows the configuration of the simulation system with smooth wall. For interatomic interactions the Lennard-Jones 12-6 potential was employed. The form of this potential is introduced in equation (2) and the interaction force between a pair is given by (3):

$$F_{ij}\left(r_{ij}\right) = -\frac{\partial V\left(r_{ij}\right)}{\partial r_{ij}} \tag{3}$$

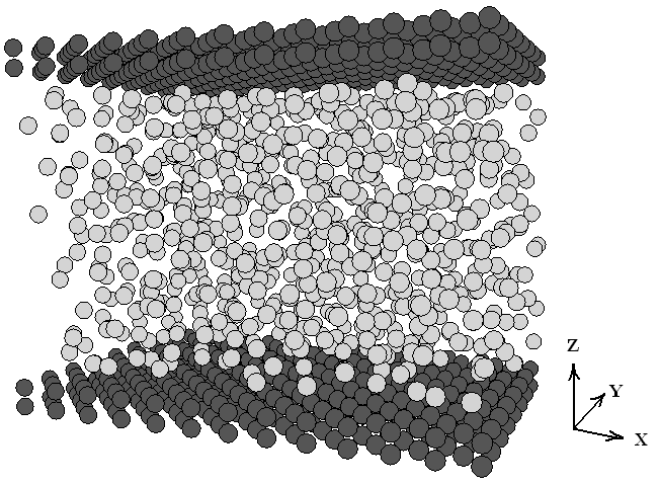


Fig.1: 3D snapshot of very early simulation of Poiseuille flow (smooth wall)

A Lennard-Jones (LJ) potential with energy scales of ε_{wf} is also used for interactions between wall and fluid atoms. To reduce the computational cost, V^{LJ} is truncated at $r_c=2.5\sigma$. The fluid temperature is initially set to $k_BT/\varepsilon=1.1$ and the density is fixed at $\rho\sigma^3=0.8$ which correspond to the liquid state of argon [24]. Liquid argon is allowed to flow in hydrophobic and hydrophilic nanochannel by varying the wall-fluid binding energy (ε_{wf}) from 0.4 to 4.0. The characteristic time ($\tau=\sigma\sqrt{m/\varepsilon}$) is 2.16×10^{-12} s, and the simulation uses 4 fs (~0.002τ) time steps [25]. Periodic boundary conditions were imposed in the streamwise (in x and y) direction as described in [26]. The system measures $h=10\sigma$ between the walls and 15.5σ in x-direction and 10.0σ in the y-direction.

For establishing the baseline for the approach the fixed lattice wall model was applied first. Each wall consists of atoms forming two [001] planes of an FCC crystal. Two different wall models were utilized. First by implementing fixed lattice wall, the wall atoms are constrained to stay in their lattice position. Since the walls were modeled as perfectly elastic fixed lattice crystals, there is no heat transfer to/from the walls [24]. Under such unrealistic thermal interface model, the temperature of the system steadily increases if any kind of work is done on the system. Therefore, to hold the wall temperature constant, a thermostat (Nose-Hoover) is necessary. In order to properly implement thermal interactions between the fluid and wall molecules, a lattice bond springs as described in [18] and [21], were utilized. The simulation is continued by applying another wall boundary condition which allows the wall atoms to move. To maintain a well-defined solid structure with a minimum number of solid atoms, each wall atoms is attached to its lattice site by a harmonic spring of constant k. The spring constant k controls the thermal roughness of the wall and its responsiveness to the fluid. k is adjusted so that the mean

squared displacement of about the lattice sites is less than the Lindemann criterion for melting. For the results shown $<\delta u^2>/d^2=0.01$ was used. By implementing thermal wall model, momentum and energy exchanges between fluid and the wall, take place properly. Therefore, excessive heat in the fluid is transferred to the walls, and then, the heat is dissipated through the thermostat applied on the walls and the wall temperature is held constant.

The fifth order predictor-corrector gear algorithm was used for time integration of MD equations. In order to find the position and velocity of molecules at the next time step, evaluation of intermolecular forces are required. Detailed forces of interactions exerted on each of the fluid molecule and wall molecules are calculated separately by using interaction potentials between fluid molecules and fluid molecules and wall molecules. The total force exerted on a fluid molecule is therefore a sum of these two terms, plus the external force:

$$F_{fluid}(r_i) = \sum_{j=1}^{N_f} \frac{\partial V\left(r_{ij}\right)_{fluid-fluid}}{\partial r_{ij}} + \sum_{j=1}^{N_w} \frac{\partial V\left(r_{ij}\right)_{fluid-wall}}{\partial r_{ij}} + F_e \qquad (4)$$

F_e is the external driving force applied in the x direction to drive the fluid atoms in the simulation of Poiseuille flow. Every fluid molecule interacts with other fluid or wall molecules within the predetermined cutoff distance. The forces as described by relations (2) and (3) depends on interparticle distances. Hence, the resulting force on a molecule of the wall is given as:

$$F_{wall}(r_i) = \sum_{j=1}^{N_f} \frac{\partial V\left(r_{ij}\right)_{wall-fluid}}{\partial r_{ij}} + K\left|r_0 - r_i\right| \qquad (5)$$

Where k is the wall crystal bonding stiffness and $| r_0 - r_i |$ is the distance between the original lattice position and current position of the wall molecule. k was chosen to be 500 which would satisfy the Lindemann criterion.

To start the finite-difference algorithm, liquid particle positions must be assigned at time t=0. The cubic structure is applied for fluid molecules. However, the cubic structure will be melting when the pairwise Lennard-Jones force acts on the liquid particles in the determined temperature. At the beginning of the calculation, randomly distributed velocities are assumed to activate the calculation. In order to make sure that the steady-state can be easily reached after some reasonable time, the randomly distributed velocities should satisfy the case that the total kinetic temperature is equal to the kinetic energy.

$$\frac{3}{2} N k_B T_{fluid} = \frac{1}{2} m \sum_{i=1}^{N_f} \left(v_{i,x}^2 + v_{i,y}^2 + v_{i,z}^2\right) \qquad (6)$$

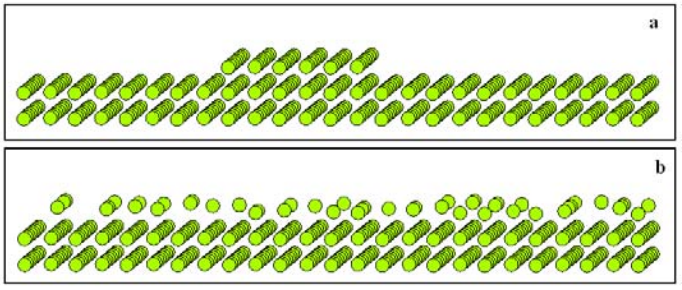
The main purpose of this paper is to examine the different effects of various surface roughnesses on flow behavior. Therefore two distinctive forms of rough wall were studied. In the first form named as "organized roughness", the wall was equipped with rows of atoms forming a nanostrip perpendicular to the flow. This nanostrip is constituted of Nr atoms and can vary in height to change the roughness amplitude. Nr indicates the number of total roughness atoms. Provided that, the surface roughness is mainly because of the different techniques employed in fabricating the nanochannel, the second form of the roughness is proposed to accommodate for a more realistic surface roughness. In the second form the roughness atoms were placed randomly on the lower wall with known roughness amplitude and Nr, while maintaining its FCC crystal. Therefore, by changing its roughness amplitude and atoms, a comparison can be made between the effects of first form of roughness (named as organized) and second form of roughness (named as disorganized). This would lead to a more realistic rough wall since the roughness atoms were randomly built on the wall.

The velocity profiles were used to quantify the dynamic behavior and the structure of fluid molecules in a steady state, with the computational domain divided into bins of width Δz along the z axis and the averages taken over time for each bin.

RESULTS AND DISCUSSION

Simple fluid was allowed to flow in smooth and rough channel with organized roughness and disorganized roughness. Wall-fluid interaction ranges from 0.4 to 4.0 while the wall speed can be 1.0 and 2.0. The roughness are characterized by its height, number of atoms and formation (organized or disorganized) which are named as R-Amp, Nr and D.O. or O. respectively. To make a comparison between the results Nr is chosen to be 16, 32 and 48 and R-Amp is selected as 1 and 2. The roughness is constructed on the lower surface, while the upper wall is always smooth.

To study the effects of disorganized roughness on the flow field, the number of roughness atoms, Nr, was set constant while the roughness amplitudes varied between 1 and 2. The different surfaces, used for an arbitrary Nr, according to various roughness amplitudes are shown in figure 2.



Surface roughness (a)Organized Roughness, Roughness Amplitude=1, Nr=48
(b)Disorganized Roughness, Roughness Amplitude=1, Nr=48

Fig. 2: 3D snapshot of different rough surfaces studied with the same Nr

It is observed in all the cases that the presence of any kind of surface roughness always suppresses the fluid slip for hydrophilic and hydrophobic surface nanochannels as shown in figure 3. Figure 3a shows the velocity profile for different surface roughness and lower wall-fluid interaction and in the figure 3b the velocity profile is illustrated for high wall-fluid interaction.

Apparent slip occurs at the solid interface in hydrophobic surface due to the weak interaction between wall and fluid , however as the roughness increases, the slip decreases as illustrated in figure3. It is also shown that the disorganized roughness has more frictional effect on the flow than the organized roughness on both hydrophobic and hydrophilic surfaces. By examining all the velocity profiles for different cases, it can be concluded that the velocity profile is highly depended on surface wettability and roughness.

Figure 4 shows the number density profiles along the z direction with various surface roughness and wall-fluid interaction condition. Density oscillation is observed in fluid layers adjacent to the wall. It is shown that regardless of the surface roughness, the density structure for the higher wall-fluid interaction bears higher oscillation in its profile.

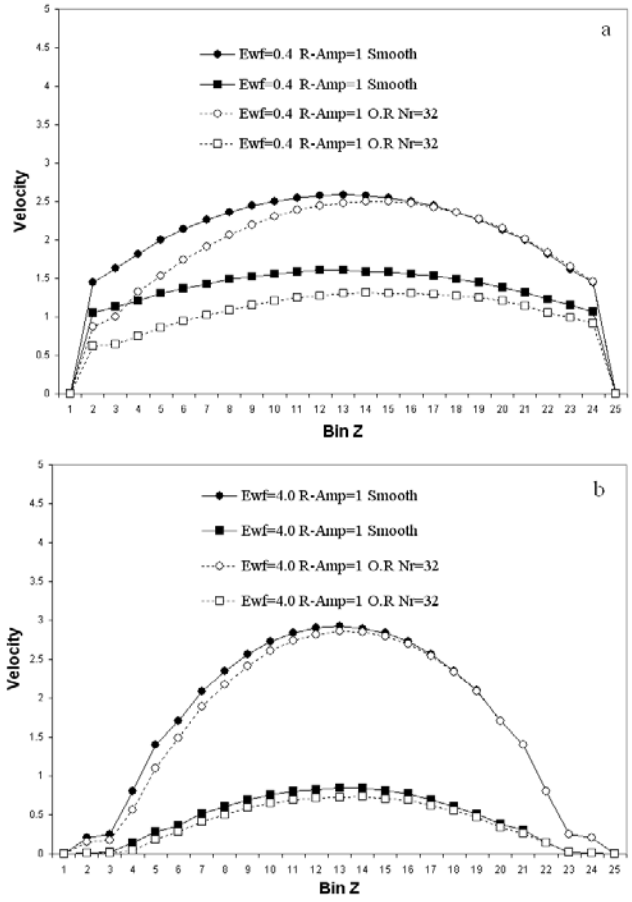


Fig. 3: Velocity profiles for different wall-fluid interaction and surface roughness

Figure 5 illustrates the variation of density profile for different organized and disorganized roughnesses and different surface wettabilities. In all cases ε_{wf} determines the structure of density profiles. Thus, density structure is highly affected from wall-fluid interaction, and more than roughness forms.

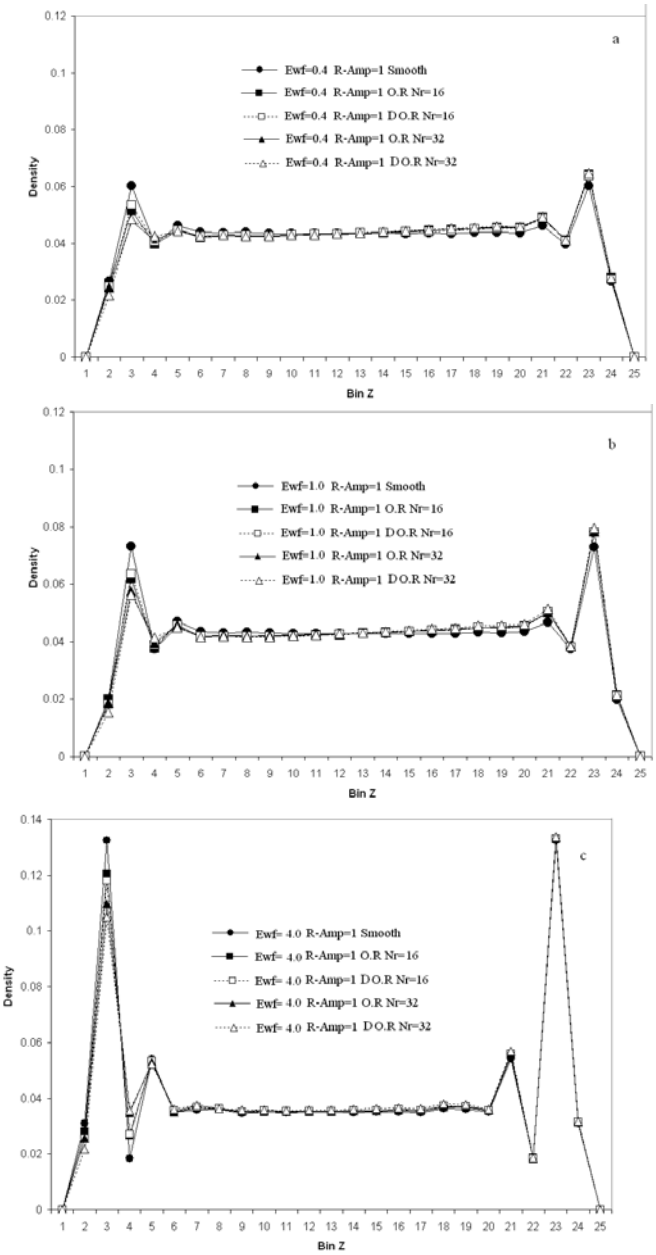


Fig. 4: Number density profiles along the z direction with various surface roughness and wall-fluid interaction condition. (O.R: Organized Roughness and DO.R: Disorganized Roughness)

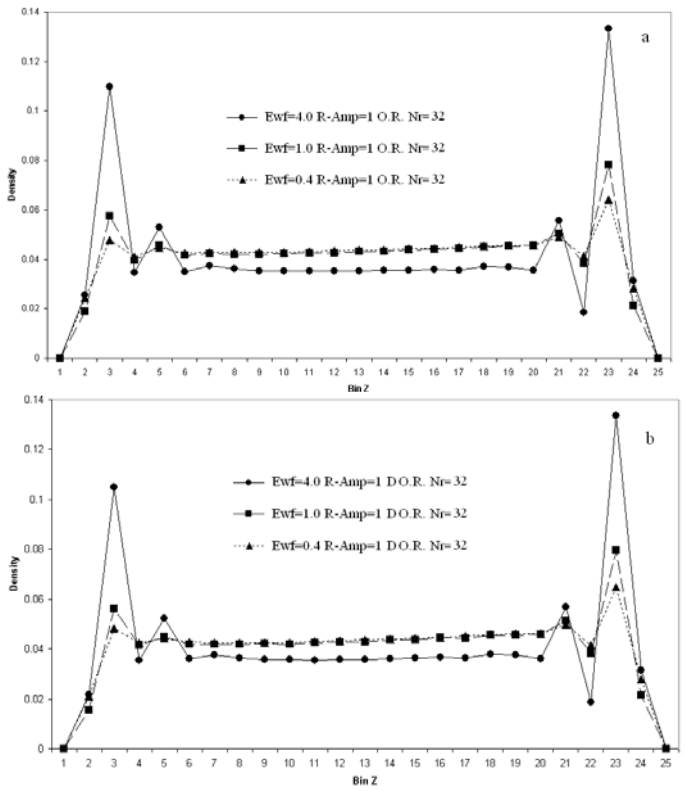
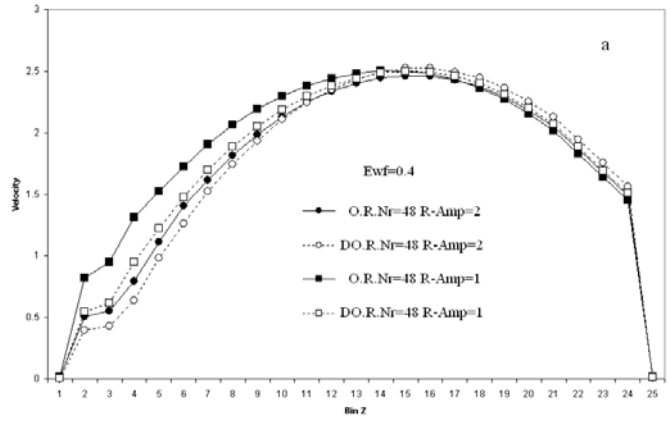


Fig. 5 : The variation of density profile for different roughness, organized and disorganized (O.R: Organized Roughness and DO.R: Disorganized Roughness)

Figure 6 shows the effects of roughness amplitude on velocity profile. It is observed that for the same Nr, the frictional effect of organized roughness with the amplitude of one is lower than all the other types of roughness with the same selection of wall-fluid interaction.



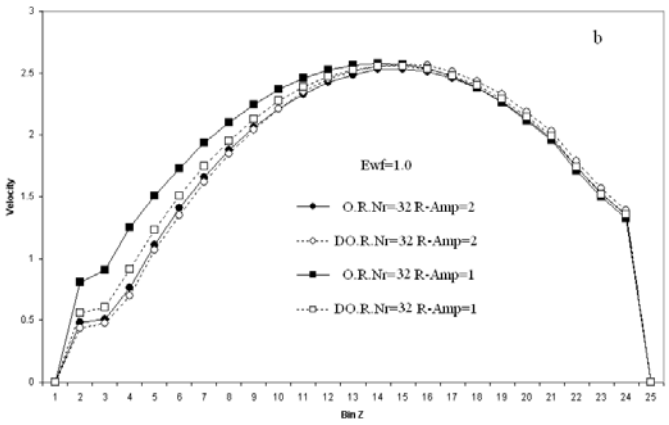


Fig. 6: The effect of Roughness Amplitude on velocity profile for various wall-fluid interactions. (O.R: Organized Roughness and DO.R: Disorganized Roughness)

CONCLUSIONS

A computer program has been developed based on the Molecular Dynamics (MD) method to perform a numerical simulation for investigating the flow behavior of simple fluids in nanochannel undergoing Poiseuille flow with different surface wettability, wall speed and roughness' forms. The solid wall was decorated with different types of roughness, Organized and Disorganized. Moreover, the amplitude of the roughness was allowed to vary along their random positioning of wall atoms that constituted disorganized surface roughness. The simulation results indicate that wall-fluid interaction and surface roughness are both important in determining the nanostructure and temperature profile of simple fluid in fluid flow in a nanochannel. It was shown that the frictional effects of disorganized roughness is higher than organized roughness.

REFERENCES

[1] U. Heinbuch and J. Fischer, (1998) "Liquid flow in pores: Slip, no-slip, or multilayer sticking" Phys. Rev. A, 40, 1144.
[2] Pit R, Hervet H, Leger L (2000) "Direct experimental evidence of slip in hexadecane: solid interfaces". Phys Rev Lett 85:980– 983
[3] Zhu Y, Granick S (2001) "Rate-dependent slip of Newtonian liquid at smooth surface". Phys Rev Lett 87:096105
[4] Bonaccurso E, Kappl M, Butt HJ (2002) "Hydrodynamic force measurements: boundary slip of water on hydrophilic surfaces and electrokinetic effects". Phys Rev Lett 88:076103
[5] Choi CH, Johan K, Westin A, Breuer KS (2003) "Apparent slip flows in hydrophilic and hydrophobic microchannels". Phys Fluids 15:2897–2902
[6] Craig VSJ, Neto C, Williams DRM (2001) "Shear-dependent boundary slip in an aqueous newtonian liquid". Phys Rev Lett 87:054504
[7] Tretheway DC, Meinhart CD (2002) "Apparent fluid slip at hydrophobic microchannel walls". Phys Fluids 14:L9–L12
[8] Cottin-Bizonne C, Charlaix E, Bocquet L, Barrat JL (2003) "Lowfriction flows of liquid at nanopatterned interfaces". Nat Mater 2:237–240
[9] Ou J, Perot B, Rothstein JP (2004) "Laminar drag reduction in microchannels using ultrahydrophobic surfaces". Phys Fluids 16:4635–4643

[10] Thompson PA, Troian SM (1997) "A general boundary condition for liquid flow at solid surfaces". Nature V389(6649):360
[11] Cieplak M, Koplik J, Banavar JR (2000) "Molecular dynamics of flows in the Knudsen regime". Phys A 287:153–160
[12] Nagayama G, Cheng P (2004) "Effects of interface wettability on microscale flow by molecular dynamics simulation". Int J Heat Mass Transfer 47:501–513
[13] Richardson S (1973) "On the no-slip boundary condition". J Fluid Mech 59:707–719
[14] C. Cotton-Bizonne, J. L. Barrat, L. Bocquet, and E. Charlaix, (2004) "Dynamics of simple liquids at heterogeneous surfaces: Molecular-dynamics simulations and hydrodynamic description". Eur. Phys. J. E 15, 427-438.
[15] D. Kim and E. Darve, (2006) "Molecular dynamics simulation of electro-osmotic flows in rough wall nanochannels" Phys. Rev. E 73, 051203.
[16] R. Qiao, (2007) "Effects of molecular level surface roughness on electroosmotic flow" Microfluid. Nanofluid. 3, 33.
[17] S. C. Yang (2006) "Effects of surface roughness and interface wettability on nanoscale flow in a nanochannel" Microfluid Nanofluid 2: 501–511.
[18] Z.Y. Guo, Z.X. Li, (2003) "Size effect on microscale single-phase flow and heat transfer", Int J. Heat Mass Transfer 46 (1) 149–159.
[19] Z.X. Li, Z.Y. Guo, (2003) "Some flow and thermal phenomena at microscale", keynote, in: 1st International Conference on Microchannels and Minichannels, Rochester, NY, USA, pp. 77–92.
[20] Moran Wang, Jinku Wang, Shiyi Chen (2007) "Roughness and cavitations effects on electro-osmotic flows in rough microchannels using the lattice Poisson–Boltzmann methods" Journal of Computational Physics 226, 836–851.
[21] A. Kharazmi, R. Kamali (2009) "Molecular Dynamic Simulation of surface roughness effects on nanoscale flows" 7th International ASME Conference on Nanochannels, Microchannels and Minichannels, June 22-24, Pohang, South Korea.
[22] D. Rodriguez-Rius, F.J. Garcia_Saban (2005) "Physico-chemical characterization of the surface of 9 dental implants with 3 different surface treatments" Med Oral Patol Oral Cir Bucal;10:58-65.
[23] Wennerberg A, Albrektsson T, Lausmaa J. (1996) "Torque and histomorphometric evaluation of c.p. titanium screws blasted with 25- and 75-um-sized particles of Al_2O_3". J Biomed Mater Res 1996;30:251-60.
[24] Thompson, PA. & Robbins, MO. (1990) "Shear flow near solids: epitaxial order and flow boundary conditions". Phys Rev A, 41(V41),6830.
[25] Kim, B.H.,Beskok, A. & Cagin, T. (2008) "Thermal interactions in nanoscale fluid flow: molecular dynamics simulations with solid-liquid interfaces". Microfluidc Nanofluidic , 0267,7.
[26] Allen, MP. & Tildesley, DJ. (1989) "Computer simulation of liquids", Oxford University Press, Oxford.
*[27] Barrat J-L, Chiaruttini F (2003), "Kapitza resistance at the liquid–solid interface". Mol Phys, **V101**(N11):1605–1610.*