# Surface Roughness and Hydrodynamic Boundary Slip of a Newtonian Fluid in a Completely Wetting System

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The influence of surface roughness on the boundary condition for the flow of a Newtonian fluid near a hard wall has been investigated by measurement of the hydrodynamic drainage force. The degree of slip is found to increase with surface roughness. This leads to the conclusion that in most practical situations boundary slip takes place, leading to a reduction of the drainage force.

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In order to apply the Navier-Stokes equations to a fluid flowing over a solid surface it is usually assumed that the liquid molecules adjacent to the solid are stationary relative to the solid. This is known as the no-slip boundary condition and has been successfully applied to model many macroscopic experiments for more than a hundred years, although there are conditions where this assumption leads to unrealistic behavior. Recently, sensitive measurements on confined Newtonian liquids have revealed that partial slip is occurring at the boundary [1–5]. This complicates description of flow in these systems, as knowledge of the degree of boundary slip is required.

Control of the boundary condition will allow a degree of control over the hydrodynamic forces in confined systems and be important in lubrication and microfluidics. It has been demonstrated that the degree of slip increases both with the viscosity of the solution and the shear rate [3,5]. It is also widely expected that wetting will affect the slip behavior [6,7]. The cohesive force at the interface is important in determining the degree of slip, as this interaction must be overcome (if only momentarily) in order for slip to occur. Similarly, the cohesive forces between a solid and a liquid determine the wetting behavior. Therefore it has long been recognized that in a system that exhibits boundary slip the degree of slip will be related to the wettability [8]. In reference to water, a hydrophobic surface is expected to give rise to a partial-slip boundary condition [9] and a completely hydrophilic surface is expected to give rise to a no-slip boundary condition. Indeed, the original experiments of Whetham [10] that led to the acceptance of the no-slip boundary condition were made on hydrophilic surfaces. Recently it has been demonstrated that slip occurs even on completely wetting surfaces [2].

Another important surface characteristic in determining boundary slip behavior is the roughness. Few surfaces are smooth on a molecular scale and therefore knowledge of the relationship between surface roughness and the degree of boundary slip is important and is the subject of this study. On an atomic scale, roughness will influence the number of neighboring surface atoms with which an interfacial solvent molecule can interact and on a slightly larger scale will alter the pattern of liquid flow. This had led to predictions that surface roughness will both increase the degree of slip at a surface [11,12] and alternatively lead to a no-slip boundary condition [13,14]. Recently, Pit *et al.* [4] and Zhu and Granick [15] have demonstrated that in the presence of polymer films, which increase the surface roughness, the degree of boundary slip is reduced. We demonstrate here that in a completely wetting system (advancing contact angle 0°), in the absence of polymer, the degree of slip *increases* as the surface roughness increases.

The hydrodynamic drainage force on a borosilicate glass sphere (radius $12.55 \pm 0.1\ \mu\text{m}$) approaching a wall (silicon wafer) perpendicularly in a viscous liquid has been measured using an atomic force microscope (Nanoscope III, Veeco). The method is similar to that described previously [3]. The sphere is attached to a fine cantilever spring ($0.15\ \text{N}\,\text{m}^{-1}$) [16] following the method of Ducker *et al.* [17] and Butt [18] enabling the drag force to be quantified. The approach rate and surface separation is accurately controlled using a piezoelectric transducer and corrected for the deflection of the cantilever spring. The deformation of the microsphere in contact with the substrate has been estimated using Johnson-Kendall-Roberts theory [19] to be less than 0.1 nm. Aqueous solutions of sucrose of varying concentration whose temperature has been equilibrated at 20 °C before each measurement are used as Newtonian viscous liquids.

We compare the experimental results to the existing theoretical models. The first is an approximate calculation for the no-slip boundary condition; the hydrodynamic force exerted on a sphere of radius $r$ approaching a flat surface perpendicularly with a velocity $V$ is given $F_h = 6\pi\eta r V \frac{r}{h}$, where $\eta$ is the bulk dynamic viscosity and $h$ is the surface separation. The error in the approximation can be determined using the exact calculation of Brenner [20] and is insignificant when $h$ is much smaller than $r$, a condition satisfied in this system ($h/r < 0.07$). The second model is a calculation by Vinogradova [9,21] allowing for slip at the solid boundary. In this model the hydrodynamic drag force exerted on a sphere approaching a flat surface at a distance $h$ is $F_h = \frac{6\pi r^2 \eta V}{h} f^*$, where $f^*$ is the correction factor for slip for two surfaces with the same slip length $b$:

$$f^* = \frac{h}{3b}\left[\left(1 + \frac{h}{6b}\right)\ln\left(1 + \frac{6b}{h}\right) - 1\right].$$

The theory of Vinogradova can also be applied to surfaces with dissimilar slip properties [22]. This model is accurate for low Reynolds numbers and for small surface separations ($h \ll r$). Both approximations are valid for our measurements. In using the no-slip model there are no adjustable parameters, as all the parameters (sphere radius, liquid viscosity, and spring constant) are known by independent measurement. In using the Vinogradova model the surface slip length is adjusted to give the best fit to the experimental data.

Sucrose solutions were used as Newtonian viscosity standards. The clean silicon wafer is extremely smooth (0.7 nm rms). Following treatment with 30% aqueous KOH at 70 °C for between 20 and 450 s the roughness of the silicon surface can be increased [23] and determined by imaging the surface with an atomic force microscope (AFM). All roughness measurements were made over an area of 2 $\mu$m × 2 $\mu$m. The surface is oxidized ($SiO_2$) and is completely wetted by water. Figure 1 shows AFM images of both native and chemically roughened silicon wafers (0.7, 4.0, and 12.2 nm rms) used in the experiments reported here. Please note that all surfaces are "mirrorlike" and are, in comparison to the majority of surfaces, very smooth.

The hydrodynamic force as a function of surface separation is presented for the three surfaces and compared to the theoretical no-slip force in Fig. 2. Note the actual approach velocity of the surfaces is altered by the deflection of the cantilever and must be determined from the experimental data. This results in a degree of experimental noise being manifest in the theoretical plot in this figure. This is described more fully elsewhere [16]. Clearly slip is present in all cases. This confirms the presence of boundary slip on completely wetting surfaces [2]. For roughened surfaces the repulsive force generated on approach is as little as half of the theoretical no-slip value ($f^* = 1$) and is also considerably reduced from that of the very smooth surface. Clearly, roughened surfaces experience considerably diminished hydrodynamic forces in comparison to model smooth surfaces that are employed in most studies.

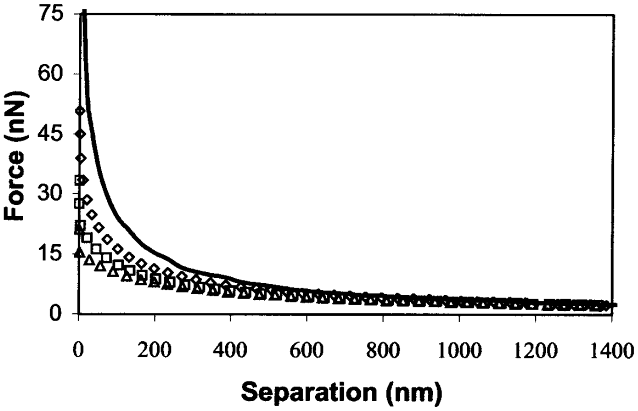

FIG. 2. Measured hydrodynamic drainage force versus separation for three surfaces [roughness decreases from the bottom curve up, 12.2 nm (triangles), 4.0 nm (squares), 0.7 nm (diamonds) rms] is compared with the no-slip theoretical drainage force (top curve) (drive rate 32.4 $\mu$m s$^{-1}$, viscosity 39.00 mP s). As the roughness increases, the magnitude of the hydrodynamic force decreases. Only every tenth data point is shown for clarity.

In order to aid comparison between measurements made under differing viscosity and velocity conditions and to aid in the determination of boundary slip, plots

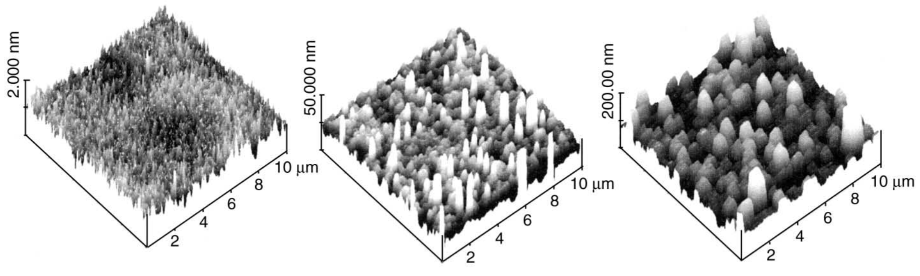

FIG. 1. Atomic force microscope images of silicon wafer substrates roughened by treatment with KOH, obtained in contact mode; image area is 10 $\mu$m × 10 $\mu$m. Treatment for 0, 50, and 120 s (from left to right), has resulted in surfaces of rms roughness 0.7 nm [ ~ 1.8 nm peak to peak (PP)], 4.0 nm ( ~ 20 nm PP), and 12.2 nm ( ~ 50 nm PP), respectively. Note the change in vertical scale between images (2, 50, and 200 nm from left to right). The roughness features are a result of etching along the crystallographic planes of the silicon wafer. The same three surfaces are used throughout.

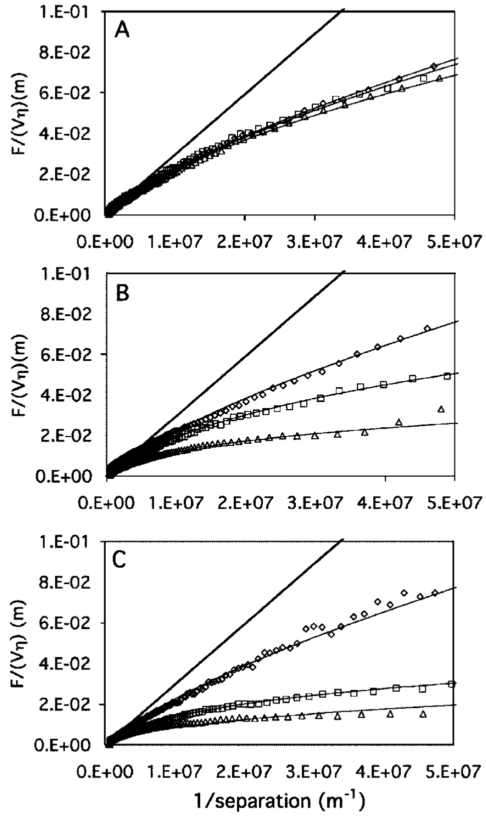

FIG. 3. $F_h/(V\eta)$ vs 1 separation for three different sets of experimental conditions. A substrate drive rate of 16.2 $\mu$m s$^{-1}$ in an aqueous sucrose solution of viscosity 6.15 mP s (39.98 wt%) (a); drive rate 16.2 $\mu$m s$^{-1}$, viscosity 19.22 mP s (51.94 wt%) (b); and drive rate 43.2 $\mu$m s$^{-1}$, viscosity 6.15 mP s (c). Measurements are shown for surfaces with a range of rms roughness [0.7 nm (diamonds), 4.0 nm (squares), 12.2 nm (triangles)]. The data are seen to deviate from linearity indicating the presence of a partial slip boundary condition. The more pronounced the surface roughness the larger the magnitude of the boundary slip that is observed. Table I indicates the slip lengths for the sphere and substrate that have been fitted to the data in Fig. 3.

of $F_h/(V\eta)$ versus $1/h$ are presented in Fig. 3. In this format, the no-slip theory shows a linear behavior. The experimental data, on the contrary, exhibit a nonlinear behavior and are indicative of a slip boundary condition. Attempts were made to fit the data using the same slip length and using dissimilar lengths for the sphere and substrate. Our fits with only one slip length for both surfaces shows that the slip length increases with surface roughness (Fig. 2). We find that for the smoothest substrate the best fit to our data is obtained when the slip on one surface is near zero, whereas for the rougher substrates the best fit is obtained using values for slip on both surfaces, that is the lower of the two slip values must be increased. Therefore we first set the slip on the flat surface to zero and determine the slip on the sphere. The sphere is slightly rougher than the substrate but the magnitude of the slip suggests that the macroscopic curvature of the sphere may play a role. Then we keep the value on the sphere constant during the fits on the rougher flat surfaces. Note that the degree of slip could be increasing on both surfaces but we are unable to determine the slip lengths uniquely from our data. The lowest viscosity and approach rate is employed in Fig. 3(a). In order to increase the degree of slip and therefore more clearly delineate the effect of surface roughness the viscosity of the solution or the approach rate can be increased. As the viscosity is increased the shearing force on molecules adjacent to the surface increases and surface interactions are more easily overcome. In Fig. 3(b) the viscosity has been increased to 19.22 mP s. As the surface roughness increases, the degree of slip increases and the hydrodynamic force is diminished. In comparison to Fig. 3(a) the approach velocity has been increased and the viscosity remains unchanged in Fig. 3(c). A substantial increase in slip results, and the slip length is again seen to increase with the surface roughness.

For all the results presented here, and for the theoretical calculations, we define the zero of separation using the hard contact between the surfaces obtained experimentally. The asperities will deform elastically during contact to a small degree that cannot be fully quantified. Even during contact this deformation will be insufficient to flatten the surfaces and remove the surface roughness. A rough surface can no longer be adequately described as an infinitely sharp boundary, as the interface has considerable depth. This complicates the definition of zero separation between the surfaces and our analysis. Should this zero be defined at the peaks, or the valleys, or at an intermediate position? Experimentally, the surfaces come

TABLE I. Slip lengths fitted to the data in Fig. 3. All lengths are in nanometers.

| System | Fig. 3(a) | | Fig. 3(b) | | | Fig. 3(c) | | |
|---|---|---|---|---|---|---|---|---|
| Substrate roughness rms (nm) | Sphere | Substrate | Sphere | Substrate | Both | Sphere | Substrate | Both |
| 0.7 | 43 | 0 | 44 | 0 | | 42 | 0 | |
| 4.0 | 43 | 1 | 44 | 20 | | 42 | 135 | 80[a] |
| 12.2 | 43 | 3.5 | 44 | 225 | 100[a] | 42 | 900 | 175[a] |

[a] It is not possible to distinguish between an equal slip or different values of slip on the sphere and the substrate from the data in these instances; therefore the values show the bounds of the possible slip lengths. The true slip length is likely to be greater on the sphere and less on the substrate than the values shown.

into contact when the peaks make contact and we use this contact to define zero separation. The question arises as to whether this is the appropriate zero of separation for fluid flow. Our experiments are not sensitive enough for this to be determined. However, the degree of slip and the differences in the degree of slip between the surfaces of different roughness are sufficient that the variation in the measured hydrodynamic forces cannot be attributed to an error in the zero separation due to surface roughness.

We note that previous reports have found that the presence of polymer films increases the surface roughness and inhibits slip [4,15]. In comparison, the surfaces employed in this study are free of polymer. Our conclusions on the effect of roughness are not incompatible with these experiments. It is widely accepted that adsorbed polymers move the plane of shear into the fluid, giving rise to a negative slip length. If the polymer surfaces employed have chains protruding into solution, the effect will be to reduce the degree of slip. This demonstrates that the measure of surface roughness employed here (rms roughness) is not adequate to fully describe the relationship between the structure of a rough surface and the degree of boundary slip. A measure of the roughness over a range of scales is likely to be required. Alternatively, the presence of polymers on the surface alters the slip behavior in another manner.

In conclusion, we have shown that surface roughness that is large on the scale of the fluid molecules significantly increases the degree of boundary slip in a completely wetting system. This has implications for the description of real world systems where the majority of surfaces will exhibit surface roughness at scales similar to and larger than the surfaces used in this study. This inevitably leads to the conclusion that a great number of systems (or problems) with liquid-surface boundary conditions are likely to exhibit boundary slip and a corresponding reduction in the hydrodynamic drainage force will result.

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