Influence of Humidity on Friction Forces in Point-Contact under Small Loads

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The frictional force in point-contact between a Si3N4 probe and a polished SUS304 steel surface was investigated by lateral force microscope under different press forces and relative humidity (RH). Variations in the coefficient of friction due to adsorbed water and meniscus necking between the probe and the steel surface were analyzed. At 65% RH, the coefficient of friction between the probe and the hydrophilic surface sharply decreased from 4.0 to 1.0 as vertical nominal load increased from 6 nN to 35 nN, slowly decreasing to 0.7 when the load was increased to 66 nN. At a vertical load of 14 nN, the coefficient of friction for a hydrophilic surface increased with humidity, peaking at 3.5 at an RH of 65%, but decreasing when RH increased to 80%. The change was analyzed to be due to the amount of adhered water on the surface and the formation of meniscus necking. The coefficient of friction on a hydrophobic surface with 1H,1H,2H,2H-perfluorodecylethoxysilane (PFDS) was much lower and independent of humidity, despite the much large adhesion force occurred between probe and PFDS molecular. (Received October 27, 2016; Accepted November 28, 2016; Published January 16, 2017)

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1. Introduction

Machines have undergone microminiaturization largely to save energy, resources and space. However, the ability of downscaled machines to work as well as large classic scale machines remains unclear, even if the former can be precisely manufactured. The frictional force between sliding objects must be carefully considered because this force can greatly influence movement and energy efficiency. In particular, clarifying details of friction associated with ultra-small point-contact (USPC) in MEMS is becoming important. Lateral force microscope (LFM), basing on atomic force microscope (AFM) technology, is currently used to measure the force or coefficient of friction under small loads. In LFM measurement, a micro cantilever twists due to the frictional force between the cantilever tip and the specimen surface, with deformation due to the twist precisely determined from the change in position of the laser reflection from the cantilever back to the photo detector.

To date, there have been many measurements and analyses of friction under small loads1–7). The frictional force can be influenced by temperature, humidity, roughness, chemical environment and test-machine dynamics3,5,6). In particular, frictional force is influenced by the adhesion of foreign materials, such as adsorbed water from ambient air, and lubrication oil on the sliding interface. However, adsorbed micro-scaled water on solid surfaces is generally difficult to be observed directly, and the effects of adsorbed micro-scaled water on the characteristics of friction between the small tip and the solid surface have not been clearly determined. In contrast, the ac non-contact mode (so as being also called as tapping mode) of AFM has been used to image micro water and oil droplets with precise control of the force of interaction between the probe tip of the cantilever and micro liquids8,9), providing a new method of investigating USPC friction and clarifying adsorption morphology. Direct observation can therefore provide knowledge of the coefficient of friction under small loads.

This study investigated the USPC friction force between a Si3N4 cantilever tip and a polished stainless steel using the LFM mode of an AFM at various applied press forces, sliding velocities, and humidity conditions. Variations in the coefficient of friction due to adsorbed water on the specimen surface and meniscus necking between the probe tip and the plane of the specimen surface were analyzed.

2. Experimental Procedure

The specimen analyzed was a commercially supplied SUS304 stainless steel sheet, which was cut into pieces measuring 20 mm x 20 mm x 2 mm and wet-polished by alumina powder of mean diameter about 50 nm. The surfaces were ultrasonically cleaned in acetone three times for 900 seconds each, using fresh acetone samples for each cleaning. In addition, a specimen of SUS304PFDS, consisting of polished steel coated with a thin evaporated layer of the organic compound 1H,1H,2H,2H-perfluorodecylethoxysilane (C16H3gF17O3Si; PFDS) was used. The thickness of the PFDS layer covering the whole steel surface was estimated as about 2 nm and the evaporation details as well as the film analysis can be found in the previous report10).

The specimen surface was viewed using both the contact mode (c-mode) and the ac non-contact mode (ac-mode) of an atomic force microscope (AFM; SPM-9700, Shimadzu Co.) in a chamber with freely controlled temperature and humidity (Fig. 1). Figure 2 shows schematics of the c-mode, the LFM mode and the ac-mode used for observations and measurements. Generally, in c-mode scanning (Fig. 2(a); ac)), the cantilever tip touched the specimen surface with a constant repulsive force (perpendicular to the specimen surface) between them; thus, soft microscopic species such as adsorbed water and organic contaminants on the surface can be displaced from the scanning area and only the hard substrate surface
can be observed\textsuperscript{8,9}. In the ac-mode (Fig. 2 (b)), however, a much smaller tapping force was applied between the cantilever tip and the surface, allowing the soft microscopic species on the surface to be observed directly\textsuperscript{8,9}. Moreover, moving the cantilever in the lateral direction in c-mode, shown as the in/out direction in Fig. 2 (a; a\textsubscript{2}), results in frictional (lateral) force between the cantilever tip and the specimen surface, also called the LFM mode. The bending displacement in c-mode and LFM mode scanning can be measured by the vertical variation in position of the reflected laser point on the photo detector, which can be used directly to determine the applied press force (or vertical nominal force). In contrast, torsional displacement of the cantilever caused by the frictional force can be detected by the lateral variation of the position of the reflected laser point on the photo detector (\(E\textsubscript{d}\)), yielding the frictional (lateral) force (Fig. 3). The coefficient of friction could also be obtained by calculating the ratio of the vertical (press) force to the lateral (frictional) force.

Two types of micro cantilever were used in this work. The first (Type A) was a Si-cantilever (Nanosensor Co., NCHR-W; typical resonance frequency \(f_r = 330\) kHz, radius of curvature of the tip \(R < 10\) nm), which could be used in both the c-mode and the ac-mode, but mainly in the latter. The second (Type B) was a Si\textsubscript{3}N\textsubscript{4}-cantilever (Olympus Co., OMCL-RC800PSA), which could be used in the c-mode and the LFM mode. The dimension of the chip with Si\textsubscript{3}N\textsubscript{4}-cantilevers (Type B) is shown in Fig. 4; only the longer upside chip was used in this work. The dimensions and mechanical constants of the cantilevers given by the maker of Olympus Co. are shown in Table 1. Before measuring the frictional force, a referential Al\textsubscript{2}O\textsubscript{3} ceramics block was used to generate a force curve of the AFM (Fig. 5). Many parameters, including (i) the relationship among the vertical nominal force, adhesion force and the deflection of the cantilever and (ii) the reflection properties of the laser light in the apparatus, could be obtained from the force curve. During the scanning of the cantilever on the specimen surface, the operating point (\(OP\)) of the tip was varied from 1.0 \(\sim\) 8.0, corresponding to the change in reflected laser point position \([\text{A + B} - \text{(C + D)}]\) on the photo detector (Fig. 1), which was proportional to the applied vertical press force \(F_N\) between the lever tip and the specimen.

The frictional force between the sharp Si\textsubscript{3}N\textsubscript{4} probe and the stainless steel was measured in the LFM mode in response to vertical nominal forces ranging from 6 nN to 66 nN with RH ranging from 25% to 80%. The temperature in the chamber was pre-set at 296 \(\pm\) 2 K. The sliding velocities of the probe on the specimen surface were altered from 2.5 to 40 \(\mu\text{m/s}\) with the sliding distance of 5 \(\mu\text{m}\). To determine the effects of

![Fig. 1](image1) Environmental atomic force microscope (AFM) used for surface observations and to measure friction and adhesion forces.

![Fig. 2](image2) Schematic of (a) contact mode (c-mode) and lateral force microscope (LFM) mode, (b) ac non-contact mode (ac-mode) of the atomic force microscope (AFM).

![Fig. 3](image3) Principal of measuring friction force with the laser lever method. (a) Scanning directions of the cantilever, (b) output of laser deflection on photo detector, (c) torsion of the cantilever and laser position on the photo detector.

![Fig. 4](image4) Dimensions of the cantilever used in lateral force microscope (LFM) mode (Type B).
adsorbed water on the frictional force, the specimen surface was monitored using both the c-mode and the ac-mode of AFM with the Type A cantilever to determine the thickness of the water layer. In all these experiments, RH in the chamber was gradually increased from 20% to its target value and maintained for 900 s before assessments. Finally, a theoretical analysis was performed to determine the intrinsic essence of the micro friction. In addition, the adhesion force was determined by measuring the pull-off force of the cantilever, obtained from a force curve, on the specimen surface.

3. Results and Discussions

Figure 6 shows the nominal coefficient of friction ($\mu_0$) between the $\text{Si}_3\text{N}_4$ tip and the SUS304 surface at 65% RH at different sliding velocities, with $\mu_0$ calculated from eq. (1):

$$\mu_0 = \frac{F_L}{F_N},$$

where $F_L$ is the frictional force obtained by measuring the torsional deformation of the cantilever and $F_N$ is the press force applied by controlling the bending deformation of the cantilever. The surface of SUS304 was hydrophilic, with a contact angle of 20 degrees, for millimeter scaled water droplets. Each $\mu_0$ value indicates the average of six tests on different lines randomly chosen on the substrate, with the bars showing the standard error. The large standard errors in the case of 6 $\mu$m correspond to the much smaller friction force and the not so changed S/N noise ratio in the measurement. Alterations in sliding velocity from 2.5 to 40 $\mu$m/s had no effect on the coefficient of friction. This might be due to all the much slow sliding velocities in this experiment and the not appeared lubrication between the probe tip and the steel by water. In contrast, increasing the applied press force from 6 to 66 nN reduced the coefficient of friction. Figure 7 (a) shows a redrawn curve of coefficient of friction vs. applied press force at a sliding velocity of 10 $\mu$m/s. The coefficient of friction was much higher, near 4.0, at a press force of 6 nN, decreased sharply to 1.0 at a press force of 35 nN, and then gradually decreased to 0.7 at a press force of 66 nN. These findings were similar to those obtained on mica and pure nickel surfaces at 30% RH. In general, the coefficient of friction between a ceramic and a metal, with a much larger area contact and press force, is less than 1.0, indicating that the coefficient of friction will not change much even at a press force much higher than 66 nN.

Figure 8 (a) shows the coefficient of friction between $\text{Si}_3\text{N}_4$ (tip)/SUS304 (plane) at a sliding velocity of 10 $\mu$m/s and a
constant press force of 14 nN under different RH. The coefficient of friction increased as RH increased, peaking at about 3.5 at an RH of 65% and decreasing thereafter. Note that the coefficient of friction, 3.5, was higher than that in Fig. 6, although they should be the same because the conditions were identical. This difference may have been due to the use of an older probe, which generates a relatively larger lateral force.

Figure 8 (b) shows the coefficient of friction on the surface of SUS304PFDS as a function of RH. The surface of SUS304PFDS was hydrophobic, with a contact angle of 114 degrees for a millimeter-scaled water droplet. The coefficient of friction was much lower than that of the hydrophilic SUS304 and was almost constant, at about 1.2, regardless of RH.

4. Discussion

The coefficient of friction is determined by both the frictional force and the press force. As shown in Fig. 6, Fig. 7 (a) and Fig. 8 (a), (b), only the applied nominal force was used as the press force to determine the coefficient of friction. However, the actual press force when water is adsorbed on the surface should have included an additional adhesion force (pull-off force; $F_{ad}$) from a meniscus cross-linking the tip of the cantilever and the plane of the steel. The thickness of adsorbed water will influence the shape of the meniscus cross-linking and the adhesion force. Adsorbed water can generally promote the physical and chemical stabilization of the surface, which can be observed in the ac-mode of AFM.

4.1 Thickness of adsorbed water

Figure 9 shows the typical procedure used to assess adsorbed water on the hydrophilic SUS304 surface and the measurement of average water thickness by scanning the surface in the ac-mode and the c-mode. In the ac-mode of AFM, the Type A cantilever was used. The ac-mode showed particle-like objects on the surface (Fig. 9 (c)). However, c-mode observation of the central area (Fig. 9 (d)) showed that most of these particles had disappeared, indicating that the particles in Fig. 9 (c) are due to soft adsorption of water perhaps also including some impurities. Re-observation by ac-mode after c-mode scanning showed the replaced water from the central area (Fig. 9 (e)), indicating that the adsorbed water on the surface included particles/droplets and film-like objects.

The amount of water can be determined from the ac-mode image after scanning with c-mode, by calculating the three-dimensional volume of regions B and C (Fig. 10). The lowest base point was present on c-mode scanning of area B after the removal of water. The volume was divided by the total area to determine the mean height of surfaces B and C ($t_B$ and $t_C$), with $t = t_C - t_B$ being considered as the mean thickness of adhered water. Figure 11(a) shows the effect of RH on mean
water thickness on the hydrophilic surface. Thickness increased monotonically from 2.2 to 5.8 nm as RH increased from 35% to 80% RH. Figure 11(b) shows the calculated thickness of adhered water between the probe tip and the plane surface of $t$. A comparison of the calculated (Fig. 11(b)) and measured (Fig. 11(a)) thickness of water showed that both tended to increase as RH increased, but that measured thickness was greater than calculated thickness, which was likely due to the rougher surface of the measured ($R_{a} = 32$ nm) than the ideal plane used in the calculation. The calculated relation of $t-h_{0}$ shown in Fig. 11(b) was obtained by using the eqs. (4), (5) and (6) described in section 4.2.

In contrast, the measured water thickness on the hydrophobic surface (Fig. 11(c)) remained almost constant, at about 2 nm, indicating that almost no water was absorbed on the hydrophobic surface even after RH was increased. The detected “water thickness” on this surface may only be the thickness of the PFDS molecular film, which has a long chain of about 2 nm$^{10}$.

### 4.2 Adhesion force between the tip and the plane

A water meniscus neck generally forms between the spherical tip and the plane surface in ambient air, as shown in the inset to Fig. 11. The curvature radius ($R$) of the tip of the probe (Type B) is about 20 nm in measuring frictional force and force curve. The adhesion force in this section has been described$^{14}$. Since the tip touches the plane substrate during friction scanning, the distance $a$ shown in this figure should be zero. However, according to the consideration of the physical explanation in the case of 0% RH and the calculation of van der Waals force, it was set as 0.2 nm here. The adsorbed water has wetting contact angles of $\theta_{1}/\theta_{2}$ calculated relation of 60 degrees to the tip and 20 degrees to the SUS304 plane. Thus the curvature radii of the meniscus water on the tip side ($r_{1}$) and both sides ($r_{2}$) can be calculated as in eqs. (2) and (3).

$$r_{1} = R \sin \phi$$  

$$r_{2} = \frac{a + (R - R \cos \phi)}{\cos(\theta_{1} + \theta_{2}) + \cos \theta_{2}}$$  

The thickness of the water meniscus ($t$) can be expressed as in eq. (4), which can be also treated as the thickness of the adsorbed water on the plane surface.

$$t = a + R(1 - \cos \phi)$$  

Using the Laplace (eq. (5)) and Kelvin (eq. (6)) equations, the relations of $\varphi - p/p_{f}$ and $p/p_{f} - t$ can be built.

$$\frac{\Delta p}{\gamma} = \frac{1}{r_{1}} - \frac{1}{r_{2}} = \frac{1}{R \sin \phi} - \frac{\cos(\theta_{1} + \theta_{2})}{a + R(1 - \cos \phi)}$$  

$$\kappa T \ln \frac{p}{p_{f}} = \frac{\Delta p}{\gamma}$$

Here, $\Delta p$ is the difference in pressure between the inside and outside of the meniscus due to the surface curvature of the meniscus, $\gamma$ is the surface energy of water (73 mJ/m$^{2}$), $p/p_{f}$ is the relative humidity, $\nu_0$ is the molecular volume (0.03 nm$^{3}$)$^{9,14}$, $k$ is Boltzmann’s constant ($1.38066 \times 10^{-23}$ J/K) and $T$ is temperature (K).

Theoretically, the adhesion force $F_{ad}$ is the sum of four terms (eq. (7)):

$$F_{ad} = F_{C} + F_{vdW} + F_{E} + F_{B},$$

where $F_{C}$ is the capillary force including surface tension ($F_{S}$, as calculated in eq. (8)) and the capillary pressure force ($F_{p}$ as calculated in eq. (9)) due to the pressure difference $\Delta p$.

$$F_{S} = 2\nu R \sin \phi \sin(\theta_{1} + \phi)$$

$$F_{p} = -\nu R \Delta p = \pi \nu R \left( -\sin \phi + \frac{\cos(\theta_{1} + \phi) + \cos \theta_{2}}{R} \right)$$

$F_{vdW}$ is the van der Waals force, expressed as in eq. (10):

$$F_{vdW} = F_{water} \left[ 1 - \frac{1}{[1 + R(1 - \cos \phi)/a]^2} \right]$$

$$+ F_{air} \left[ 1 - \frac{1}{[1 + R(1 - \cos \phi)/a]^2} \right],$$

where $F_{water}$ and $F_{air}$ are the van der Waals forces of Si$_{3}$N$_{4}$/steel in air and in water, respectively, set at 28 nN and 5 nN, respectively, using data for Si$_{3}$N$_{4}$/SiO$_{2}$$^{14}$. $F_{B}$ is the electrostatic force and the $F_{io}$ is the chemical bonding force, both of which can be ignored in this work.

Figure 12 shows the measured and calculated adhesion forces between the cantilever tip and the hydrophilic planes at 35% to 80% RH, as determined by measuring the pull-off

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![Fig. 11](image1.png)

**Fig. 11** Effects of relative humidity on the thickness of adhered water determined from observation and theoretical calculations.

![Fig. 12](image2.png)

**Fig. 12** Effects of relative humidity on adhesion force obtained from force curves and theoretical calculations.
force in the force curve of the cantilever. The adhesion force on this surface (Fig. 12 (a)) increased from 35% RH to a peak of 19 nN at 65% RH and then gradually decreased thereafter as RH increased to 80%. Although these increases and decreases in adhesion force were relatively small, their tendency was almost the same as that of the frictional force (coefficient of friction) at a constant press force of 14 nN (Fig. 8 (a)). In contrast, theoretical adhesion force on the hydrophilic SUS304, including capillary pressure force, surface tension and van der Waals force, can be calculated using eqs. (2)−(10) (Fig. 12 (b)). This force gradually increased as RH increased to 95% and then decreased. Of these forces, the capillary force $F_p$ was much higher than the others. At low humidity, too little adsorbed water is available to form a meniscus between the tip of probe and the plane surface, making the adhesion force small (Fig. 12: left region of A). As humidity increases, so does the amount of adsorbed water, forming a water meniscus and producing the adhesion force that is mainly determined by the Laplace equation (eq. (4); (Fig. 12: middle region of B). Thus, by considering Fig. 13, the capillary force $F_p$ could be determined by curvature radii $r_1$ and $r_2$. As RH increases to 95%, the rate of increase of $r_1$ is greater than that of $r_2$, resulting in an increase in $F_p$. At RH > 95%, the rate of increase of $r_1$ becomes slower than that of $r_2$, resulting in a reduction in adhesion force (Fig. 12: right region of C). The reason for the difference between turning points for calculated and measured adhesion forces, at 95% and 65% RH, respectively, is unclear, but may be related to the difference between the much rougher measured surface and the smoother ideal smooth plane, resulting in a difference in contact area between the cantilever tip and the surface. Another reason may be related to the different structure of adsorbed water layer from the bulk water$^{15-17}$. The calculated forces $F_p$, $F_S$, and $F_{vdw}$ and their totals are shown in Fig. 14, which shows the turning points at high humidity for $F_p$ and $F_S$, as well as the continuous reduction in $F_{vdw}$ with the increase in humidity. These results are similar to those previously reported$^{14}$.

The adhesion force on the hydrophobic surface was much greater (Fig. 12 (c)) and was likely due to the molecular (van der Waals) force between the probe and the PFDS molecules since almost no water is adsorbed. In light of the flexible PFDS layer, the real contact area between the probe tip and the hydrophobic surface should be larger than that on the hydrophilic one, which should also have largely increased the adhesion force (molecular force). The adhesion force on the hydrophobic surface was almost completely independent of humidity since only little water can be adsorbed (Fig. 11 (c)).

### 4.3 Recalculated coefficient of friction

As described above, the real press force should include both the applied force ($F_N$) and the adhesion force (pull-off force; $F_{ad}$). Therefore, the real coefficient of friction ($\mu$) can be expressed as:

$$
\mu = \frac{F_L}{(F_N + F_{ad})}
$$

The average adhesion force $F_{ad}$ at 65% RH was found to be 19 nN, as determined from the force curve (Fig. 5). The recalculated real coefficient of friction is shown in Fig. 7 (b) as a dashed line. Compared with the nominal coefficient of friction ($\mu_0$; solid line), the gradient of the real coefficient of friction was much smaller, changing from 1.0 to 0.5 as the nominal press force increased from 6 nN to 66 nN. The slight reduction in coefficient of friction may have been due to the rough surface.

Figure 8 (a), (a) shows the dependence of coefficient of friction on RH under a press force of 14 nN for a hydrophilic surface, before and after applying the measured adhesion force to the nominal press force. The real coefficient of friction was almost unchanged and near 1.0, indicating that the analysis of pull-off force was correct. This result may also be explained by findings showing that the larger adhesion force at 65% RH resulted in a larger frictional force. In contrast, the adhesion force on the hydrophobic SUS304PFDS, due only to the van der Waals force, was much higher than that on the hydrophilic surface (Fig. 12 (c)). Moreover, the nominal friction on the hydrophobic surface is much lower than that on the hydrophilic surface (Fig. 8 (b)). Therefore, as shown in Fig. 8 (b), the real coefficient of friction was much smaller than that on the hydrophilic surface. The value of real coefficient of friction was also independent on humidity because there is only little water can be adsorbed on the hydrophobic surface even at high humidity. This result also indicates that the large lubrication effect of PFDS molecular on the steel surface.

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**Fig. 13** Effects of relative humidity on the curvature radii of $r_1$ and $r_2$.

**Fig. 14** Effects of relative humidity on calculated $F_p$, $F_S$, and $F_{vdw}$ and their total.
In this work, only the dynamic friction force under small load was measured by using the atomic force microscope technology. In addition, it should also be possible to measure the static friction force by focusing on the initial torsion of the probe in its moving. This will be investigated in the near future.

5. Conclusion

The frictional force in ultra-small point-contact between a Si$_3$N$_4$ cantilever tip and a polished stainless steel surface was investigated using the LFM mode in an AFM apparatus as a function of press forces, sliding velocities and humidity conditions. The variation in coefficient of friction was altered by the adsorbed water on the surface and the meniscus necking between the probe tip and the flat specimen surface. These findings yielded the following conclusions:

(1) The coefficient of friction between the Si$_3$N$_4$ tip and the hydrophilic SUS304 surface at 65% RH sharply decreased from 4.0 to 1.0 as the vertical nominal load increased from 6 nN to 35 nN, slowly decreasing to 0.7 when the vertical nominal load was increased to 66 nN. In contrast, sliding velocity from 2.5–40 μm/s had no effect on the coefficient of friction.

(2) At a vertical nominal load of 14 nN, the coefficient of friction between the tip and the hydrophilic SUS304 surface increased slowly with increasing humidity, peaking at 65% RH, but then dropped as RH increased to 80%. These changes were likely due to the amount of adhered water on the surface and the formation of meniscus necking.

(3) The coefficient of friction on the hydrophobic surface with PFDS was much lower than that on the hydrophilic surface and independent of humidity, which is a result of the large lubrication effect of PFDS molecular with a large adhesion force and little water adsorption.

REFERENCES