Sliding friction
*From microscopic contacts to Amontons’ law*
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The Onset of Sliding Friction

6.1 Abstract

When two objects are in contact, the force necessary to induce frictional motion is higher than the force necessary to keep the motion going. This is because, generally, the static friction coefficient increases with rest time and hence becomes different from the dynamic one. Many questions about the onset of sliding friction remain, especially since the real contact area between the objects is notoriously difficult to determine experimentally. Due to surface roughness, this area is smaller than the apparent contact area. Here we investigate the onset of sliding friction of polymer spheres on glass by using novel molecular probes in frictional contacts that reveal the local level of confinement through their fluorescence intensity. This allows us to directly measure not only the size but also the quality of the real contact area. Changes in the latter influence the friction force per unit contact area. We find that the real contact area increases with time due to bulk creep rather than creep of the individual asperities. The quality of the real contact area also increases with time due to the gradual densification of the glassy polymer surface. This surface densification is reversed at the onset of sliding and therefore responsible for the difference between static and dynamic friction.

6.2 Introduction

Friction is responsible for 30% of the world energy consumption[1]. The onset of frictional motion is critical to processes ranging from earthquake dynamics[2] to tire grip[3]. However, it remains unclear exactly how objects in contact start to move with respect to each other, for example during the commonly observed stick-slip phenomenon. During this ubiquitous type of
motion, the objects first ‘stick’ to each other, during which elastic deformation energy is stored, and next ‘slip’, giving rise to the frictional dissipation of this energy. Earthquakes are the result of such stick-slip motion of the earth’s crust along fault lines[2]. On a smaller scale, the stick-slip motion of shoes on the ground during a basketball match or the stick-slip motion of a bow on a violin string[4] result in well-known sounds. The general requirement for stick-slip to occur is that the frictional resistance drops at the onset of sliding motion[5]; in addition, the frictional system must not be too stiff to suppress deviations from stable sliding[6, 7, 2]. Most of the times, a drop in the resistance at the onset of sliding is indeed observed because the static friction force that needs to be overcome to initiate sliding increases at rest: the static friction coefficient of e.g. glass, plastic, paper, rock, wood, metal and silicon interfaces increases as the logarithm of the waiting time[8, 7, 9, 10, 11, 12]. Given the usual interpretation of Amontons’ law of friction, which states that the frictional force is proportional to the real area of contact between the two surfaces, such frictional ageing is attributed to growth of the real contact area driven by creep deformation of the surface roughness[13, 14]. During sliding, this process is then reversed because existing contact points are destroyed while new ones are added; this reverses the frictional ageing and causes the dynamic friction to be lower than the static one. Experimental evidence in support of this is however limited[8, 15, 5], because imaging the real contact area is experimentally challenging: one has to observe an interface involving a wide range of length scales. Furthermore, the mechanism that causes the strengthening may not be universal; atomic-force microscopy (AFM) experiments have shown that the formation of chemical bonds across a frictional silica-silica interface can also cause a logarithmic increase in static friction with contact time[11]; in this case it is the number of chemical bonds that is increasing, and therefore the ‘quality’ of the contact increases rather than the area involved. Without a precise measurement of the real contact area and a thorough understanding of the contact and frictional mechanics, the distinction between the quality of the contact and an increase in contact area remains difficult to make.

6.3 Experiment

In this Chapter, we combine friction tests with direct imaging of the real contact area with molecular resolution in the direction normal to the contact area and show that the difference between static and dynamic friction is
6.3 Experiment

Figure 6.1: Experimental setup and contact image. (a), A rheometer is placed on an inverted microscope. A 600 μm polystyrene (PS) sphere is glued to the rheometer tool off-center and brought into contact with a microscope cover slip. The forces on the contact are controlled by the rheometer through rotation of the tool (tangential friction force $F$) and vertical translation of the tool (normal force $N$). A monolayer of rigidochromic molecules is covalently bonded to the surface of the glass cover slip. We excite the molecules with 488 nm laser light; those molecules that are confined by the contact will emit an increased intensity of fluorescent light. (b), The inverted microscope captures the fluorescent light emitted at the real contact area. Image size is 130 μm × 130 μm, the normal force is 578 mN. To extract the total contact area from the experimental images, we apply an Otsu threshold[16] and multiply the number of high-intensity pixels with the pixel area.

not controlled by the size of the real contact area, but by the ‘quality’ of the contact: contacts that have aged longer contribute more to the friction coefficient than what would be expected on the basis of their contact area alone.

We measure the real contact area by employing rigidochromic molecules that emit a fluorescence signal that depends on their degree of confinement[17, 18]; we show here that this allows for a measurement of the contact area and the local level of confinement within that same area. We covalently bond a monolayer of such rigidochromic molecules to one of the two frictional surfaces: a microscopy cover slip inserted into our inverted microscope. On top of the microscope, we mount a rheometer with which we bring a 600 μm polystyrene (PS) sphere into contact with the functionalized cover
slip; the rheometer can move the sphere while simultaneously measuring both normal (load) and tangential (frictional) forces on the contact (Figure 6.1a). To study the contact area and confinement, the sphere-on-glass contact is illuminated from below; 488 nm laser light excites the monolayer of rigidochromic molecules on the glass surface. Only molecules that are confined by real contact between sphere and glass will emit an increased fluorescence intensity[17, 19]. The fluorescence image of the contact taken with the microscope therefore reveals the real contact area measured with molecular resolution along the normal ($z$-direction) and diffraction-limited resolution in the in-plane directions (Figure 6.1b). This real contact area is fully controlled by the roughness and deformation of the PS sphere since the glass surface is much smoother and harder[19]. To calculate the real contact area based on the fluorescence images, we set an intensity threshold that separates the contact from the background and multiply the number of contact pixels with the pixel area.

### 6.4 Results

In the experiments, we impose a chosen normal force by mechanically lowering the rheometer so that the sphere attached to the rheometer tool touches the glass cover slip. At a normal force of 400 mN, the real contact area between the PS sphere and the glass cover slip is roughly $1.6 \cdot 10^{-9} \text{ m}^2$, giving an average contact pressure of 250 MPa. At such high pressures, the PS sphere deforms elasto-plastically[20]; previously we analyzed the PS-on-glass contact mechanics in detail by comparing fluorescence and AFM imaging of the contact area with Discrete Element Method (DEM) simulations; this allowed us to show that the in-plane resolution with which the microscope records the contact is sufficient to resolve the real contact area because smaller-scale roughness on the sphere is plastically flattened [19]. Here we show that this also allows us to disentangle the quality and quantity of contact; the latter is simply given by all pixels that show an intensity that is significantly above the background, whereas the former is indicated by the fluorescence intensity of each pixel area: variations in fluorescence intensity within the contact area reveal variations in the degree to which rigidochromic molecules are confined locally (see Appendix B).

To understand how the real contact area couples to friction, we image the fluorescence while imposing a constant sliding speed of $1 \mu\text{m}/\text{s}$ on the contact over a total distance of roughly 13 $\mu\text{m}$. The resulting images (Figure 6.2)
Figure 6.2: The real contact area during sliding. The images correspond to the data presented in Figure 6.3 acquired at the indicated values of the displacement $D$. During sliding, a gradual change in contact intensity is observed, while the contact area and structure are unchanged. Images are $90 \mu m \times 90 \mu m$.

reveal roughly the same pattern reflecting the surface roughness of the sphere. However, looking in more detail, we see that the contacts change in both position and intensity: old contacts are broken and new ones are made, which do not necessarily have the same intensity. We find the behaviour of the friction force to be generic: it increases up to the static limit at which the contact breaks and the sphere starts to slide over the substrate with a lower friction force (Figure 6.3). Remarkably, this change from static to dynamic friction does not happen over a molecular length scale but rather over the typical length scale of the asperities. Comparing the evolution of the fluorescence of the contact to that of the friction force now reveals the physics behind the difference between static and dynamic friction. The fluorescence images recorded during slip show that, while the friction force decreases, the real contact area actually increases a little (Figures 6.2 and 6.3). This is surprising as the frictional force and contact area are usually assumed to be strictly proportional to each other. We must therefore conclude that it is not
just the size of the contact area that is important but also the quality: this quality of the contact, *i.e.*, the frictional force per unit contact area apparently decreases during sliding[21]. The rigidochromic molecules provide the first direct observation of this quality-of-contact effect: the total fluorescence signal closely follows the behaviour of the frictional force. As the fluorescence signal is a quantitative measure of the confinement (*i.e.*, the free volume) of the rigidochromic molecules (see Appendix B), this means that during the renewal of the contacts at the onset of sliding the sphere surface must become less compact, and that this is the reason why the frictional force drops. These observations were made many times, and do not depend sensitively on the material used or the surface roughness.

![Figure 6.3: Friction, real contact area and fluorescence intensity during sliding. The static friction force is the peak friction value, the dynamic friction force is the steady state friction value measured during slip. The fluorescence intensity is obtained by integrating the fluorescence intensity measured in the microscopy images and subtracting the background fluorescence. All data are normalized to their end values.](image)

How does this tie in with the general observations on frictional ageing? Without sliding, the static friction force is known to increase logarithmically with time, indicating that the contact also evolves when a normal but no tangential force is applied. To track this evolution in time, we look at how the details of the contact area evolve in sliding experiments after an increasing waiting time, and perform experiments similar to those reported in Figures 6.2 and 6.3 but leave on the normal force for different times in order to allow the contact to age. As shown in Figure 6.4, we find that indeed the
6.4 Results

Figure 6.4: Frictional ageing. (a), Consecutive slip events recorded during one hour of contact ageing at a constant normal force of 400 mN. Images (90 µm × 90 µm) of the contact area recorded before the slip events at 30 and 3034 s are displayed as upper and lower insets, respectively. (b), Static and dynamic friction force from (a) as a function of time. The real contact area measured before each slip event evolves proportionally to the dynamic friction force; the fluorescence intensity measured before each slip event evolves proportionally to the static friction force. The fluorescence intensity and real contact area were rescaled onto the friction axis by multiplication with a constant. The inset shows the difference between the first and last contact area recorded in the slide-hold-slide sequence (Image size 90 µm × 90 µm).

static friction force evolves roughly logarithmically with time, but also that the dynamic friction increases in a similar way. The evolution of the dynamic friction force with time scales perfectly with that of the real contact area: the increase in dynamic friction with time is a consequence of the growing contact area. The static friction coefficient however, grows faster than the contact area, and is therefore also subject to the quality of the contact. As before, we can quantify this through the fluorescence intensity: at rest the sphere surface ages into a more compact state that reduces the free volume available to the rigidochromic molecules and increases the static friction.

The increase in real contact area during ageing (without sliding) is usually attributed to creep deformation of asperities in contact. Through the fluorescence images of the contact area, we can track the growth in real contact area...
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Figure 6.5: Contact ageing. (a), A contact like that displayed in Figure 6.1b is held at fixed normal force for 3000 s. During this time we image the real contact area and observe a linear growth with the logarithm of contact age. (b), We visualize the contact area growth by subtracting the image taken 30 s after the contact was created from that taken 3000 s after the contact was created. Before subtracting the images we binarize them using an otsu[16] threshold and align them such that the difference is minimal. The growth of contact area is concentrated at the perimeter of the contact, indicating that it is the slow flattening of the PS sphere due to creep flow that drives the contact area growth. Image size is 90 µm × 90 µm.

with time. We subtract the binarized fluorescence images recorded before the first and last slip event presented in Figure 6.4b to visualize the growth in real contact area. Surprisingly, the growth in contact area mainly takes place at the perimeter of the contact. This demonstrates that the growth in contact area is in fact not driven by creep deformation of the asperities, but by creep deformation of the bulk sphere material; as the sphere slowly flattens, new contacts are created at the perimeter of the contact area. This process is accelerated by the tangential force that is imposed during friction measurements: when a normal but no tangential force is applied for the full 3000 s, we observe a smaller increase in contact area, directly proportional to the logarithm of time (Figure 6.5).
6.5 Conclusion

In conclusion, we have shown that the time dependence of friction is controlled by both the size and quality of the real contact area. This contact area is initially formed by elasto-plastic deformation of the roughness and can subsequently change due to creep deformation of bulk material. The quality of contact, or friction per unit contact area, is set by the density of the glassy PS material at the contact area and increases at rest. This process is reversed when a sufficient tangential force is applied and the contact slips. These results have potential relevance for the study of earthquakes and landslides in which the slip weakening of glassy interfaces plays a central role.
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