The Effects of Run-In on Rubber Friction

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Synopsis

Experimental measurement of the change in coefficient of friction with sliding distance of a carbon-black-reinforced rubber on either glass or Perspex surfaces increases monotonically to a constant value as required by Saibel's theory. However, this general stochastic model does not allow for a fully satisfactory physical interpretation of the effects of run-in on rubber friction. The present measurements for rubber on a glass surface agree well with those of Roth and co-workers reported many years ago. The observation of these effects on Perspex does not appear to have been reported previously. It is found that a material, probably stearic acid or zinc stearate, is deposited from the rubber onto a glass surface when the rubber slides on it.

INTRODUCTION

The general features of rubber friction have been discussed at length in several reviews[1-3] and books.[4,5] The effect of run-in has received relatively little attention, although many years ago Roth, Driscoll, and Holt[6] demonstrated its importance for rubbers sliding on glass tracks. In most cases there was a large increase in the coefficient of friction from $\mu \sim 0.5$ up to about $\mu = 4.0$ with sliding distance. However, with a slow sliding speed a decrease in the coefficient of friction was observed. An important point noted by Roth and co-workers was the transition from static to a steady rate of sliding extended over relatively large distances, up to about 10 cm, which is nearly 10 times the diameter of the circular disc specimens. Schallamach[1] rather dismissed the importance of such effects with the comment that the effect is negligible on abrasive tracks and is therefore probably due to a conditioning of the rubber surface. The possible physical or chemical changes that occur in the surface of the rubber are not identified. However, smooth tracks are often used for model studies in order to elucidate the mechanism of rubber friction and the effects due to run-in or rub-in will be seen to be significant. Saibel[7] has proposed a general stochastic model to account for the change of frictional force with sliding distance. The model, which has not been tested experimentally, is based on the concept that the frictional force is proportional to some average strength of a "welded" (or adhesive) junction multiplied by the number of junctions present at any instant of time.

The purpose of the present paper is to present data on the effects of run-in on the friction between rubber on both glass and Perspex surfaces. An effect which we appear to have noted for the first time is that material, extracted from the rubber, is deposited onto a glass track and forms a lubricating layer. Also it is shown that the functional relationship derived by Saibel[6] may be used to represent the increase of friction with sliding distance, but the theory does not offer a fully satisfactory explanation of the effect.

EXPERIMENTAL METHODS AND MATERIALS

The apparatus used to measure the frictional force has been described by Rawson and co-workers[8,9] (Fig. 1). The surface track, a glass microscope slide, or a strip of Perspex is mounted on the trolley. By lowering the platform pneumatically the trolley moves to the right at a constant speed, and in these experiments it was 0.6 mm·s^{-1} (6 × 10^{-4}m·s^{-1}). The frictional force F is measured from the deflection of the calibrated elastic beam using an optical lever. The normal force N is applied by putting suitable weights on the push rod, and hence the coefficient of friction, μ, can be measured, since $\mu = F/N$.

The rubber test pieces were discs 10 mm in diameter cut from a sheet 1.3 mm thick and were mounted on brass cylinders 13 mm high (Fig. 1), which were attached to the push rod.

The composition and some properties of the carbon-black-reinforced natural rubber are given in Table I. Prior to making a friction measurement, the rubber discs were washed with water, dried in an oven at 80°C, and allowed to cool in a desiccator and left overnight in the desiccator. The glass and Perspex surfaces were cleaned before making a friction measurement. A satisfactory cleaning procedure was found to be washing the surface with distilled water in an ultrasonic bath for 5 min, subsequently drying at 80°C and placing directly in a desiccator and leaving overnight. Several other procedures were used but were not so satisfactory. Using the present cleaning technique, the coefficient of friction for steel (12.7 mm diameter ball bearing) on Perspex was $\mu = 0.5 \pm 0.05$. This value agrees well with that obtained by Tabor[10,11] and co-workers for steel sliding on uncontaminated Perspex.

Ellipsometry measurements were carried out using an instrument described by Rawson and co-workers.[9] A 4 MW Helium–Neon laser was used as a light
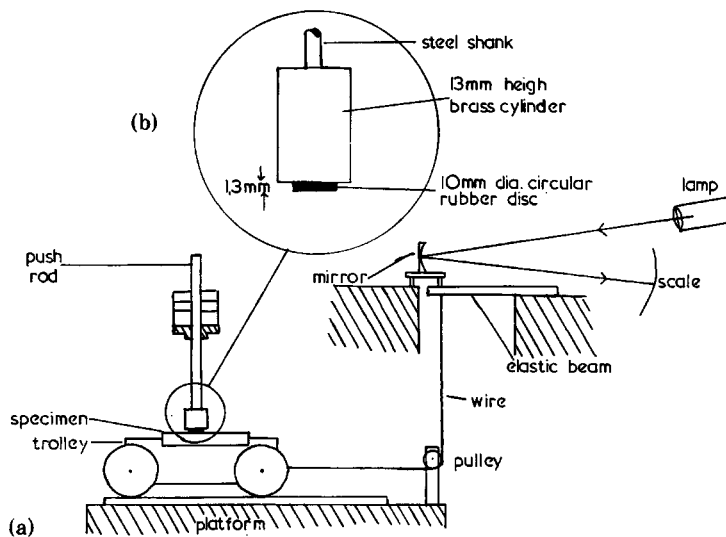

Fig. 1. (a) Schematic diagram of friction measurement; the platform moves downwards at a constant velocity and the trolley travels horizontally at constant velocity. The frictional force is measured by the deflection of the elastic beam using an optical lever. (b) Rubber disc bonded to brass cylinder.

TABLE I
Properties and Compositions of Rubber Used in Friction Tests

Properties	
Tensile strength	130 kg/cm
Elongation at break	500%
IRHD (hardness)	44
Specific gravity	1.10
Modulus 100%	8 kg/cm
200%	20 kg/cm
300%	40 kg/cm
Composition (in parts per hundred of rubber)	
SMR 5	100
Zinc oxide	5
Stearic acid	1
Octamine	2
FEF black	22
Ultrasil VN3	15
Sulphur	1.5
CBS	1
TMT	0.25

source set at a fixed angle of incidence of 70° to obtain the highest resolution, and sensitivity measurements were made of the rotation of the analyzer with and without a compensator. The interpretation of ellipsometry measurements is discussed by Vasicek.[12]

RESULTS AND DISCUSSION

The coefficient of friction vs. sliding distance for rubber on both glass and Perspex surfaces is shown in Figure 2. For the rubber sliding on glass the frictional force increased monotonically over a sliding distance of more than 4 cm. With the present apparatus the total sliding distance was limited to 6 cm, but the frictional force would not be expected to increase continuously for reasons which will become clear shortly. An increase in frictional force with distance travelled was not observed when the friction of steel on glass or Perspex was measured using the same apparatus. After initially setting the surfaces in relative motion, the friction ($\mu_{\text{steel/glass}}$ or $\mu_{\text{steel/Perspex}}$) was essentially constant for the full distance of travel allowed, that is, about 6 cm. The coefficients of friction for steel on glass or Perspex depended on the method of cleaning the surfaces prior to the measurement of frictional forces. However, with suitable cleaning procedures such as those used in the present work the coefficients of friction agreed well with those reported by Tabor and co-workers[10,11] for instance, for steel on Perspex $\mu = 0.5 \pm 0.1$; but our results were somewhat more variable than those of Tabor, with a significant variation between different samples of Perspex. Thus, it may be concluded that the increase in frictional force with distance traversed for rubber on glass or Perspex is not due to any artifact introduced by the present apparatus or method of measurement.

For rubber sliding on Perspex the coefficient of friction rises from about 1.2 to a constant value of 2.2 after sliding a distance of about 2.5 cm. Although an
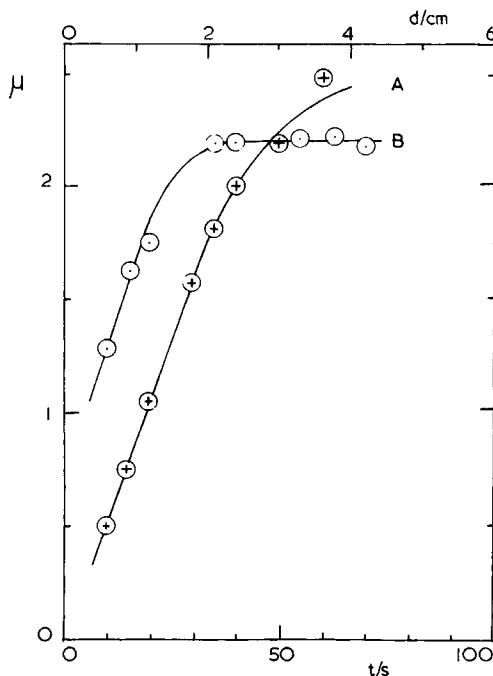
Fig. 2. Frictional force vs. distance of traverse; the force was measured 10 s after travel was initiated so that transients due to the velocity V, increasing from zero to $V_{const} = 0.06$ cm·s^{-1} were not included. Normal load, $N = 100$ g.   (A) Rubber on glass; (B) rubber on Perspex.

increase of frictional force with sliding distance has been reported previously for rubber on glass,[6] we have not traced a previous observation of this effect for rubber sliding on Perspex.

It is of interest to compare the present measurements with those of Roth et al.[6] for rubber sliding on glass (Fig. 3), and the good agreement between these results is better than might have been expected in view of the somewhat different experimental conditions and compositions of the rubber.   A new effect that we observed is also shown in Figure 3; the coefficient of friction is reduced when a rubber test piece travels along the same glass track for a second time.   This effect also occurs with the rubber sliding on a Perspex surface [Fig. 4(a)], but the decrease in friction is somewhat less than for glass.   With rubber sliding on glass [Fig. 4(b)], there is a progressive decrease in friction when the same rubber test piece slides over the same track.   Thus, either the rubber or the substrate surfaces or both are altered after the passage of the rubber test piece over either glass or Perspex.

An obvious way in which the glass surface may be altered is the deposition of a lubricating material from the rubber onto the glass.   With repeated traverses this lubricating layer is reinforced, and the coefficient of friction is further reduced.   Direct identification of the material deposited onto the surface would be very difficult, however, it has been possible to establish that a material is deposited onto the glass.   Ellipsometry measurements confirm that there is a deposit on the glass track.   There is a change in the rotation of the plane of polarized light of 3°22', which is approximately equivalent to the presence of a
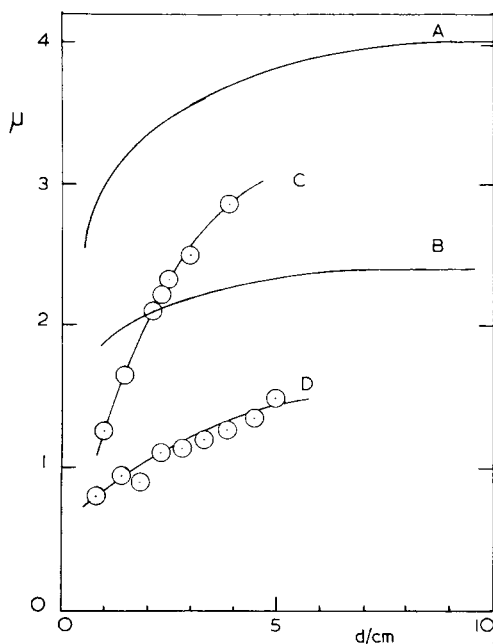
Fig. 3. Comparison of measurements of the effects of sliding distance on coefficient of friction for rubber sliding on glass. (A) Roth et al. "lightly abraded" rubber sliding on glass at 0.1 cm·s^{-1}. (B) Roth et al. rubber-abraded with 150 carborundum at a velocity of 0.1 cm·s^{-1}. *Note:* Roth et al., unfortunately, did not specify the normal pressure applied in particular experiments. (C) Present measurement—*first* traverse of cleaned rubber on glass at a velocity of 0.06 cm·s^{-1}, and normal load 100 g. (D) Present measurement—*second* traverse of cleaned rubber on glass at a velocity of 0.06 cm·s^{-1} and normal load 100 g.
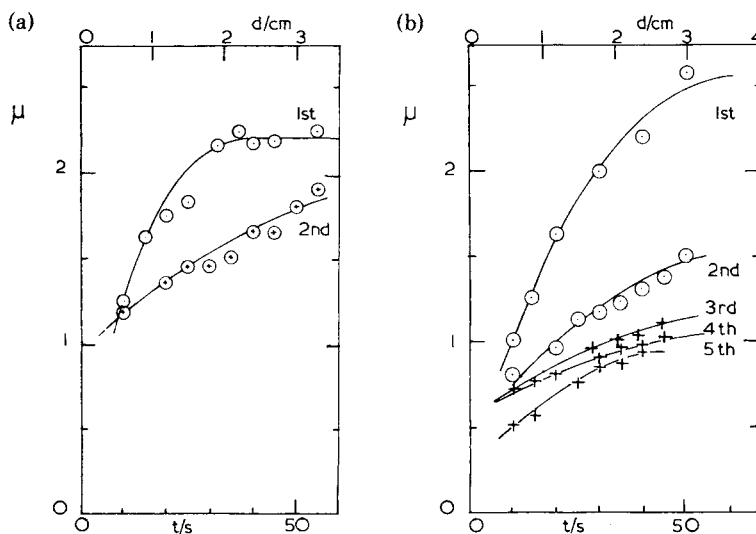

Fig. 4. Friction vs. sliding distance with conditions similar to Figure 2, and repeated traverse of the same test piece over the same track. Sliding velocity v = 0.06 cm·s^{-1}; normal load N = 100 g. (a) Rubber sliding on Perspex, first and second traverses. (b) Rubber sliding on glass—repeated traverses 1–5.

film of stearic acid (see later) of up to 100 nm thick after three traverses by the same rubber slider.

Further confirmation of this effect is provided by sliding the same test piece over fresh glass tracks; with each traverse the coefficient of friction increases (Fig. 5). After four traverses the friction became too high to measure. Also, after the first traverse the frictional force increases linearly with sliding distance over a distance of up to 5 cms. Thus, when the rubber slides on the glass, a lubricant is deposited onto the glass surface, and the concentration of the lubricant in the surface of the rubber is depleted after each traverse. Another test is that after acetone extraction of the rubber prior to a friction measurement the coefficient of friction is increased considerably (Fig. 5) and is approximately equivalent to the third traverse on a fresh track of an unextracted test piece.

Acetone extraction of the rubber has obviously removed a material that can be deposited from the rubber onto the glass during sliding. Acetone extraction will remove "soluble" materials from a sulphur-vulcanized rubber, and the possible candidates present in the original composition of the rubber (Table I) are octamine and stearic acid, if they are present after the vulcanization. It is generally accepted that acetone extraction will remove either stearic acid or zinc stearate, which is formed during vulcanization. Thus, although not fully confirmed, prime candidates for the material deposited onto the glass and probably also Perspex are stearic acid or zinc stearate.
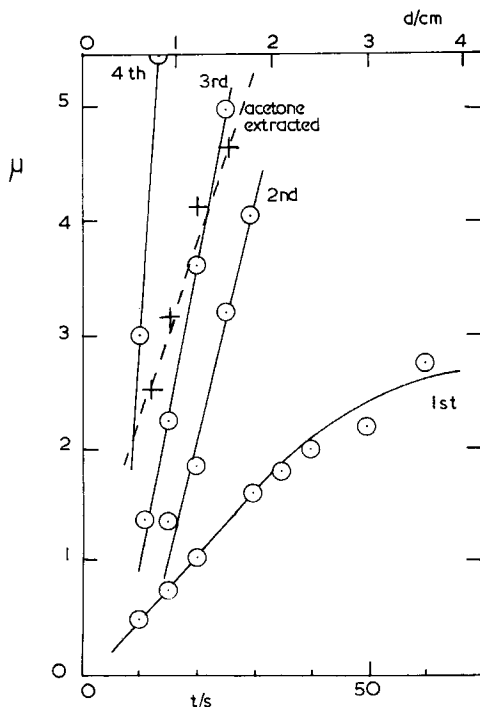

Fig. 5. Friction vs. sliding distance with conditions similar to Figure 2, and repeated traverse of the same test piece over virgin tracks. Sliding velocity v = 0.06 cm·s^{-1}; normal load N = 100 g. (————) Rubber sliding on virgin glass tracks; traverses 1–4. (- - - - - -) Acetone-extracted rubber sliding on a virgin track.

It is clear from these experiments that a material, such as stearic acid, is deposited onto glass when this rubber slides over it. However, there must also be changes in the rubber surface as well because the second traverse of a different track is not a continuation of the first track (Fig. 5). The initial absolute value of the coefficient of friction for the second traverse is lower than the limiting constant value attached in the first traverse. There are relaxation effects in the rubber surface when the load is removed and lifted from the glass. Also, there will be time for some stearic acid or other lubricating material to diffuse to the surface, and hence the coefficient of friction is lower at the start of the second traverse than the limiting constant value of the first traverse. The supply of lubricating material is limited, and hence the rate of increase of friction decreases after each removal of the rubber from the glass surface. It is obvious that further experimental work is required to elucidate these complicating effects due to changes during sliding which occur to both the rubber and the glass surfaces.

The solution deposition of a layer of stearic acid on glass provides a surface which has very different rubber friction characteristics (Fig. 6). Initially there is a small increase in the frictional force and then subsequently a decrease to a steady value of $\mu = 0.5$, which is probably about as low as can be expected for natural rubber. This behavior is quite different from the previous cases, where a material was deposited from the rubber onto the glass or Perspex. Thus, it would appear that stearic acid is being adsorbed onto the rubber surface and also possibly oriented by shearing forces. When a complete layer has been formed, the coefficient then attains a constant value.

Roth and co-workers examined the effect of water lubrication on the friction of rubber but the statement they make (Ref. 6, p. 457) is not consistent with the data they present (their Fig. 4), where it can be seen that the presence of pure water reduces the coefficient of friction marginally. It is difficult to control the thickness of a lubricating water layer which may account for the apparent contradiction. In the present measurements when the surface ahead of the sliding rubber disc was kept wet by spraying with a jet of water it was found that the friction of rubber on both glass and Perspex was significantly reduced (Fig. 6).

The increase in friction with sliding distance for rubber such as shown in Figures 2 and 3 was attributed by Kummer[13] to a rise in temperature due to heat generated by the frictional losses. However, an explicit relationship between sliding distance and friction during so-called run-in has been derived by Saibel,[7] who considered the probability of welded interfacial junctions being formed or broken. This approach is very general and is not specifically concerned with rubber friction and also does not appear to have been subjected to experimental test. In outline, Saibel considers that $N(t)$ is the number of welded junctions present at time t and $p_j(t)$ is the probability that $N(t) = j$. Also new junctions are generated at random at a constant rate ν and destroyed also at random at a constant rate η per junction so that when j junctions are operative the total extinction rate is $j\eta$. Use of a probability generating function allows the derivation of a differential equation representing the process of births and deaths, which on solution yields an expression

$$\mu(t) = \alpha \, \nu/\eta - \alpha(\nu/\eta - a)e^{-c\nu t} \tag{1}$$

where α, a, and c are constants and ν is the relative velocity between the two
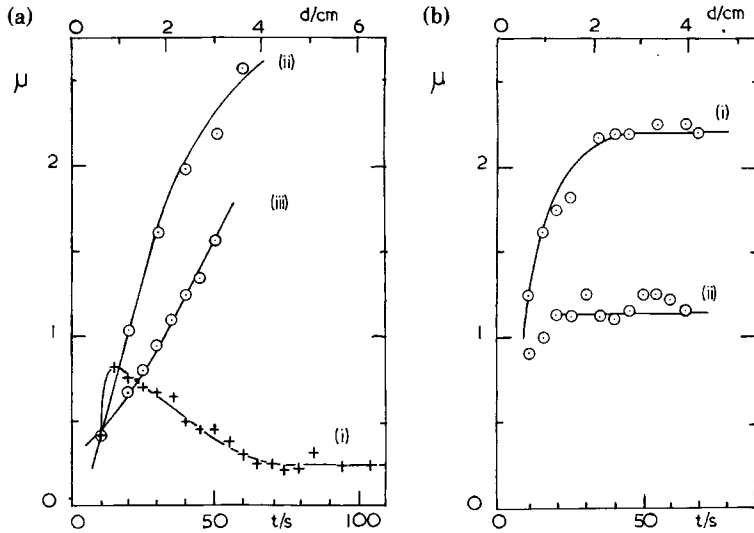
Fig. 6. Friction vs. sliding distance with conditions similar to Figure 2. Sliding velocity $v = 0.06$ cm·s^{-1}; normal load $N = 100$ g. (a) (i) Rubber sliding on glass with a solution deposited film of stearic acid. (ii) Rubber sliding on *dry* glass—as in Figure 2. (iii) Rubber sliding on glass lubricated with water. (b) (i) Rubber sliding on *dry* Perspex as in Figure 2. (ii) Rubber sliding on Perspex lubricated with water.

surfaces and t is the time. Saibel tacitly assumes that steady motion at velocity v can be instaneously attained at $t = 0$. Also the constant α is not defined. For comparison with the present data and that of Roth et al.[6] with eq. (1) it is convenient to use the following definitions

$$\mu_\infty = \alpha \, v/\eta \tag{2}$$

$$\mu_0 = \alpha a \tag{3}$$

$$cvt = bd \tag{4}$$

where d is the sliding distance and b is a constant when sliding velocity is constant (see below). With substitution of relations (2), (3), and (4) into eq. (1) the following expression is obtained:

$$\mu(t) = \mu_\infty - (\mu_\infty - \mu_0)e^{-bd} \tag{5}$$

By using

$$\Delta\mu = \mu_\infty - \mu(t) \tag{6}$$

eq. (5) can be rearranged to

$$\ln \Delta\mu = \ln(\mu_\infty - \mu_0) - bd \tag{7}$$

and hence a plot of $\ln \Delta\mu$ vs. d should be linear. The data of Roth et al. for a rubber on glass is shown in Figure 7(a) and for the present measurements for rubber on Perspex Figure 7(b).

Thus, it may be concluded that the functional form of eq. (1) is satisfactory for representation of both the measurements of Roth et al.[6] for rubber sliding on glass and the present for rubber sliding on Perspex. However, it is difficult
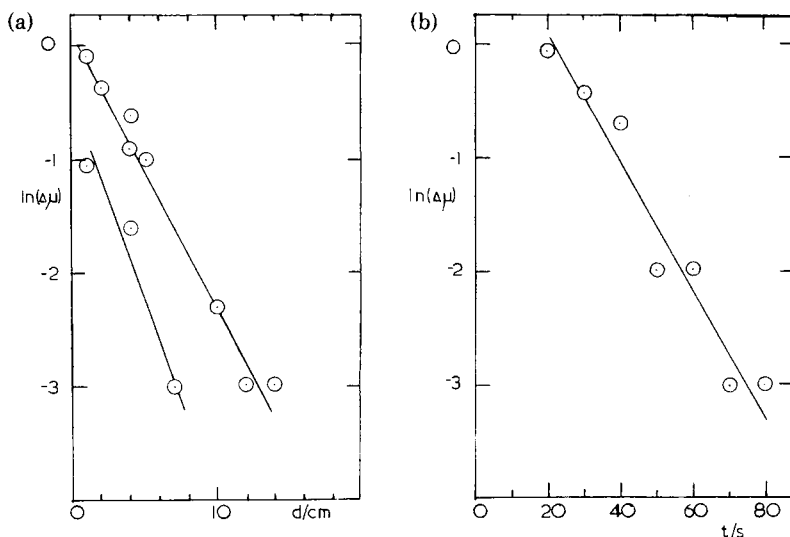
Fig. 7. Test of applicability of eq. (4). (a) Data of Roth et al.—rubber type F: (i) sliding speed 1 cm·s^{-1}; (ii) sliding speed 0.1 cm·s^{-1}. (b) Rubber sliding on Perspex; sliding speed 0.06 cm·s^{-1}.

to give a satisfactory physical interpretation of Saibel's model since α is not defined. Also from an analysis of the measurements of Roth et al.[6] we find that c in eq. (1) is not a constant independent of the velocity of sliding. Further the constant $\mu(0) = \mu_0$ for $t = 0$ and $d = 0$ should not be interpreted as equal to the static friction, a subject which has been discussed frequently. In fact, the constant $\mu(0)$ is a function of sliding speed, and, of course, the history of the two surfaces. μ_∞ is the limiting value of the coefficient of friction after a critical run-in distance has been traversed.

The increase in friction to a limiting value with distance travelled must be due to modification of the rubber surface, because on each traverse a new section of the track is moved into and hence the track cannot be modified until the slide has moved over it. The surface of the rubber may be modified by either asperities being removed, structural changes in the rubber network, or the loss of a lubricating material. It is possible that all three mechanisms are responsible for the rise in the coefficient of friction with distance travelled, but our results show that a major effect is the loss of a lubricating material from the rubber surface. If such a lubricating material is deposited on to the substrate, then with a second traverse over the same track the friction should be reduced, and this is in fact the observed behavior.

It is, of course, possible to obtain an expression equivalent to (5) simply by postulation of a first-order growth process, similar to the increase in concentration of B in a chemical reaction such as A+ $\cdots$ → B+ $\cdots$. Physical effects that may give rise to such an increase could be the setting up of a steady regimen of Schallamach waves[14,15] or the Mullins effect,[16] in which the elastic modulus of a rubber is reduced after successive stress–strain cycles. Such stress–strain cycles will also affect the viscoelastic losses and hence will cause an increase in frictional forces. In principle, it would appear that the increase in friction with sliding distance is due to an increase in real contact area between the rubber and the glass or Perspex surfaces.

The present results show clearly that the determination of the friction of rubber requires specification of the history of the two rubbing surfaces. In practical applications such as an unlubricated rubber seal in which essentially the same track is tranversed repeatedly there may well be a decrease in frictional losses until a complete layer of material such as stearic acid or zinc stearate is deposited from the rubber on to opposing surface. However, in the more general case when the rubber is always running continuously into a fresh track, the frictional force will continue to increase because the potential lubricant in the rubber surface is consumed and hence is not available for deposition on to the track. Thus, it may well happen that experimental measurements yield values for rubber friction which grossly underestimate the effective frictional losses that occur in practice.

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