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Surface Friction of Epoxidised Natural Rubber in Its Raw State

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Measurements of the sliding friction of unvulcanised samples of various grades of epoxidised natural rubber have been made against steel over a wide range of speed and temperature. Some steel tracks were deliberately roughened to find what difference this caused. The effect of polymer blending on friction and the role that plasticisers can play in reducing the surface friction were investigated in relation to practical matters.

In view of the promise of epoxidised natural rubber (ENR) as a new polymer[1], the frictional behaviour of the unvulcanised material has been investigated in a series of simple experiments aimed at an understanding of interfacial events during sliding on a steel substrate. The variation in glass transition temperature afforded by different degrees of epoxidisation bears directly upon frictional behaviour. It is of scientific interest to see whether the variation in glass transition temperature can be used as an alternative to different operating temperatures in order to construct a friction 'master-curve'. A study of the friction of ENR grades is of technological interest because it will help to foresee the process behaviour of the raw polymer in factory operations.

EXPERIMENTAL

Rubber samples were designated grades of Standard Malaysian Rubber (SMR) and ENR[1]. All were masticated down to a common Mooney viscosity of 60 ± 2. The masticated samples were compression-moulded into hemicylinders, the cavity mould surfaces being smooth and bright. Mould times were 30 min–60 min at temperatures of 110°C–120°C. Hemicylinders were left to cool in their moulds under pressure and, apart from highly epoxidised grades, they were removed without the need for release agents. For ENR-50 and ENR-70, which are moderately tacky, moulding was done against a Melinex sheet placed in the mould cavity. When these samples were to be tested the Melinex was peeled away from the cylindrical test surface. All the hemicylinders had a radius of curvature of 21 mm and axial length of 15 mm. In this raw state (no compound ingredients and unvulcanised), the hemicylinders were friction tested.

The majority of the friction measurements were made using apparatus that has been described in an earlier communication[2]. Some additional low temperature measurements were made in a deep-freeze cabinet[3]. Raw rubber hemicylinders were pulled over a smooth steel plate track under a fixed normal load of 4.5N. Sliding speeds were varied between 0.001 mms^{-1} and 100 mms^{-1} and the steel track surface temperature could be raised to 80°C by an electric hot plate placed under it. For each friction measurement, a rubber hemicylinder sample was brought into loaded contact with the steel track for at least 2 min dwell, the rubber side profile viewed and measured to find the width of the contact band, and then pulled. The contact width was most easily measured by inserting Melinex 'feelers' into either side of the contact region and recording their separation against a ruler, typical contact band widths being 7 mm to 12 mm. At temperatures of 50°C and higher, a longer dwell time of 5 min was allowed so that the rubber surface was brought to full temperature before sliding was initiated.

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RESULTS

Friction of ENR-50

Experiments were carried out with raw ENR-50 hemicylinders at temperatures of 25°C, 55°C and 80°C and data were plotted as the variation in friction coefficient, μ (friction force F/normal load W) with sliding speed *(Figure 1)*. The data show some dependence on temperature. At higher sliding speeds and lower temperatures there was a tendency to stickslip motion (*Figure 1* shows highest and lowest values of coefficient), with only slight scuffing of the rubber surface. At low sliding speeds and high temperature, motion was continuous, with material transfer to the steel track. At intermediate speeds and temperature, ridges[2] formed on the rubber surface and rolls of debris were left on the steel track.

Friction of Various ENR Grades

Friction measurements were made for different levels of sample epoxidation from zero (SMR-L) to 70 mole per cent. Despite scatter, differences in behaviour can be discerned *(Figure 2)*. In general, for a given temperature and speed, the sliding friction increases with the extent of epoxidation, and the more highly epoxidised samples had a greater tendency to stickslip motion. For low epoxidisation gross ridge formation on the sample surface often occurred, whereas at high epoxidation tiny surface cracks were observed. These cracks may reflect a lower tear strength[4].

Master-curve of Friction Data

Using the WLF equation[5] an attempt was made to transform the coefficient of friction data *(Figures 1* and *2)* into a single 'master-curve'[6]. The glass transition temperature *(Tg)* of the SMR-L sample was -70°C, and each mole per cent epoxidation raised the glass transition by one degree (all values determined by differential scanning calorimetry). Friction transforms were plotted with the experimental *Tg* values inserted into the WLF equation. The exercise was only partially successful in that the data did not pull together into a clearly defined master-curve. However, if allowance was made for the area of rubber surface observed to be in contact with the steel plate, matters improved.

The contact width made between rubber and steel was measured for every test run (see above), and from this the contact area was estimated (contact width × hemicylinder axial length). The area varied with temperature and grade of ENR. The area estimates were used to calculate friction shear stresses (friction force/contact area), and when these stresses were plotted against the reduced sliding speed $a_T V$ (a_T is the WLF transform factor) the data points were tightened into a 'master-curve', with a further improvement *(Figure 3)* coming from multiplying the shear stress by Ts/T, where $Ts = Tg + 50$, since forces in polymer chains depend upon the absolute temperature. The master-curve *(Figure 3)* shows a steady rise in friction stress with rate until the parameter $a_T V$ reached values around 4 when considerable macro-stickslip was encountered. At even higher a_T values the friction declined to a small steady value equivalent to $\mu = 0.3$, not unlike the level of friction found for plastics materials such as polystyrene, PMMA, PVC[7]. At this low level of friction all signs of stickslip motion disappeared. To reach the high $a_T V$ values meant that the ENR samples had to be cooled below 0°C, and it was found that under these conditions the sample material was hard and only suffered slight wear during sliding. The minimum temperature at which ENR-50 and ENR-70 were tested was -33°C, which implied lg $a_T V$ values as high as 39 when the sliding speed was 10 mms^{-1}. Out to such high $a_T V$ values the friction continued to remain low ($\mu \approx 0.3$) and constant for all speeds tested (0.1 – 10 mms^{-1}).

It is noted that the results presented in *Figure 3* were obtained from two distinctly different pieces of apparatus; at 25°C and above from an apparatus[2] placed on an open bench, at 0°C and below from an apparatus[3] placed in a deep-freeze cabinet. Although there was some scatter, which is to be expected, it is encouraging to see the agreement between data from the two apparatuses.

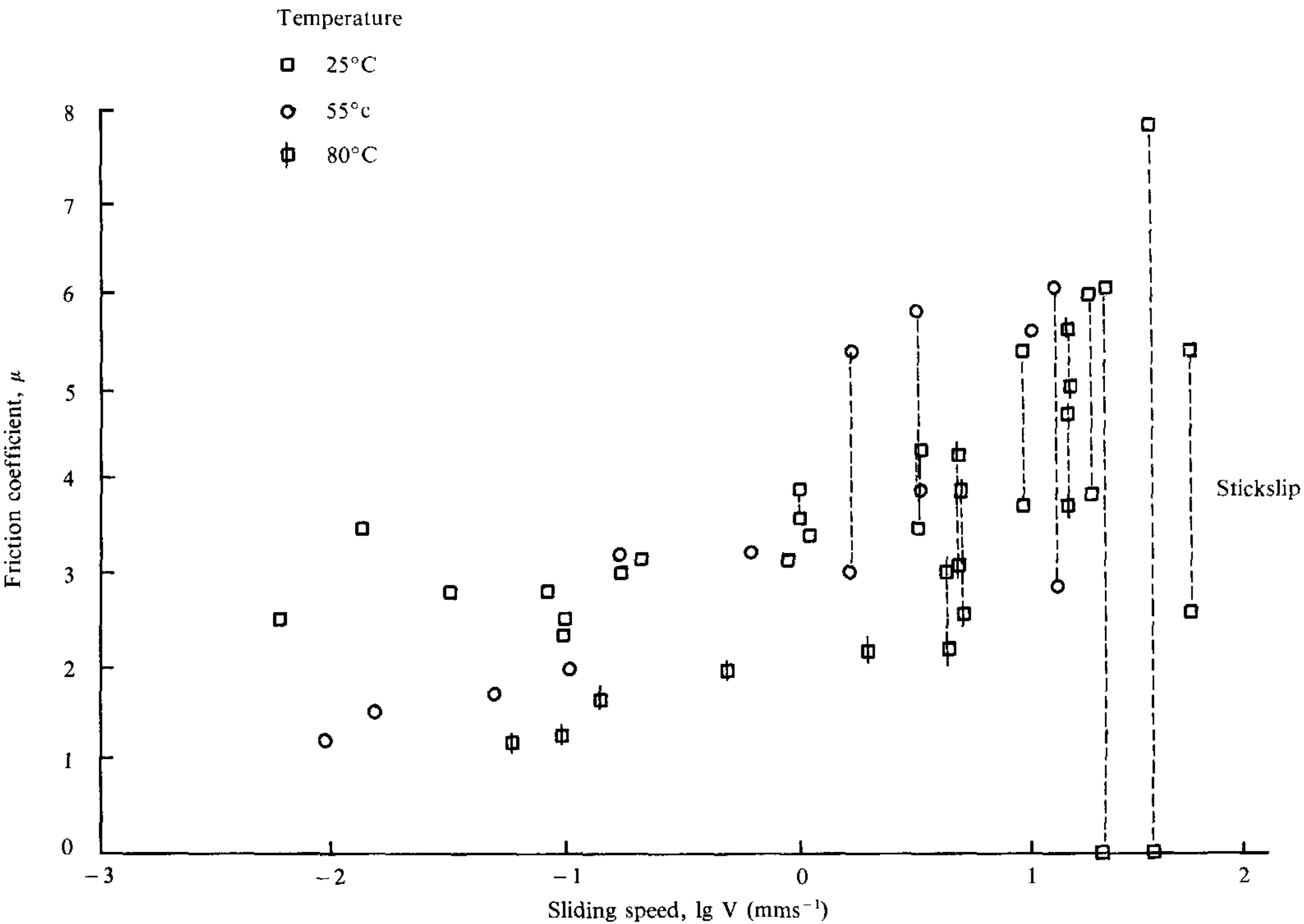

Figure 1. Friction dependence on sliding speed for hemicylinders of ENR-50 slid on a smooth steel track (0.06 μm CLA) at different temperatures. RH = 55%–65%, normal load 4.5N.

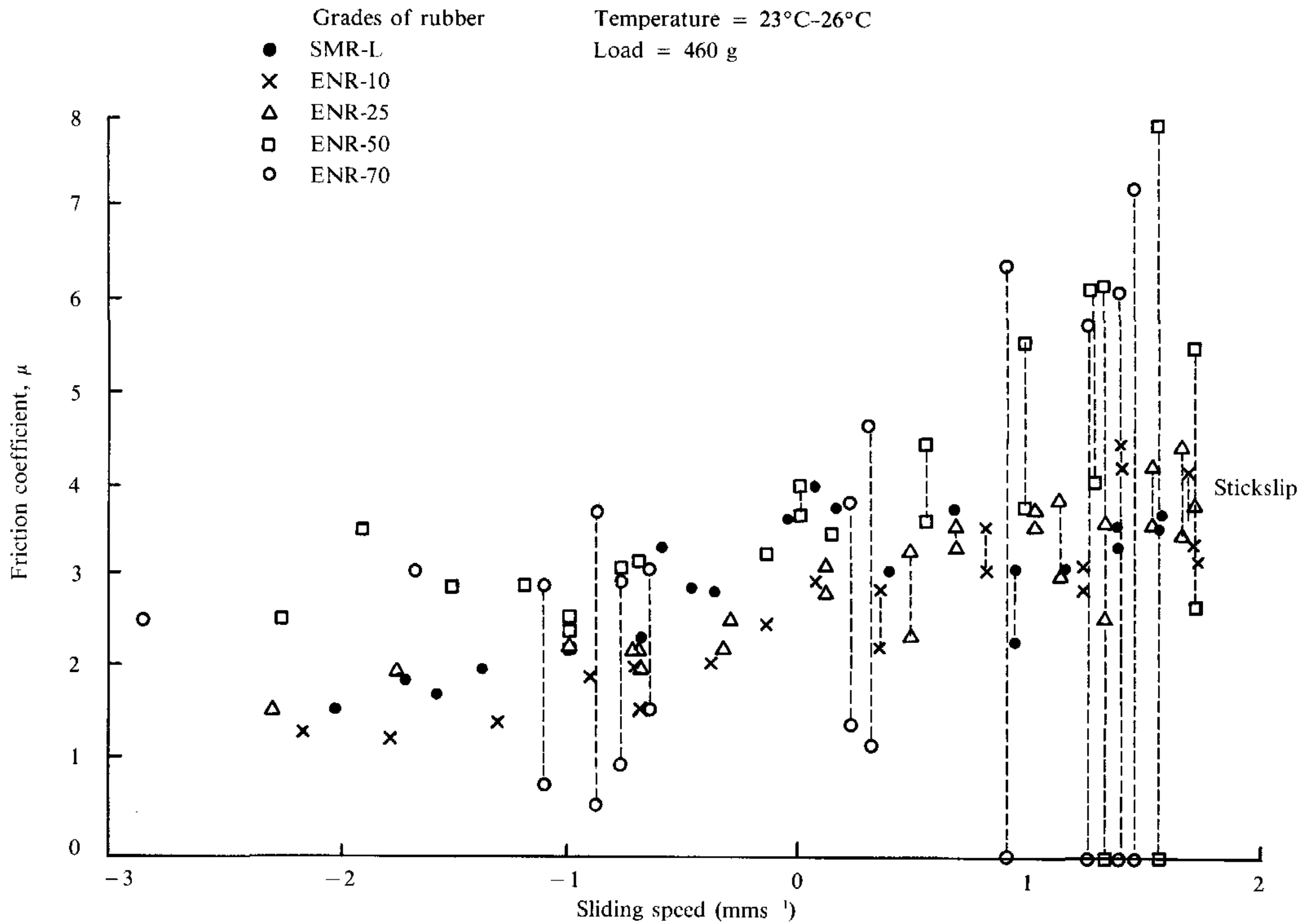

Figure 2. Friction dependence on sliding speed for hemicylinders of SMR-L and four grades of ENR slid on a smooth steel track (0.06 μm CLA). Temperature = 23°C–26°C, normal load 4.5N, RH = 55%–65%.

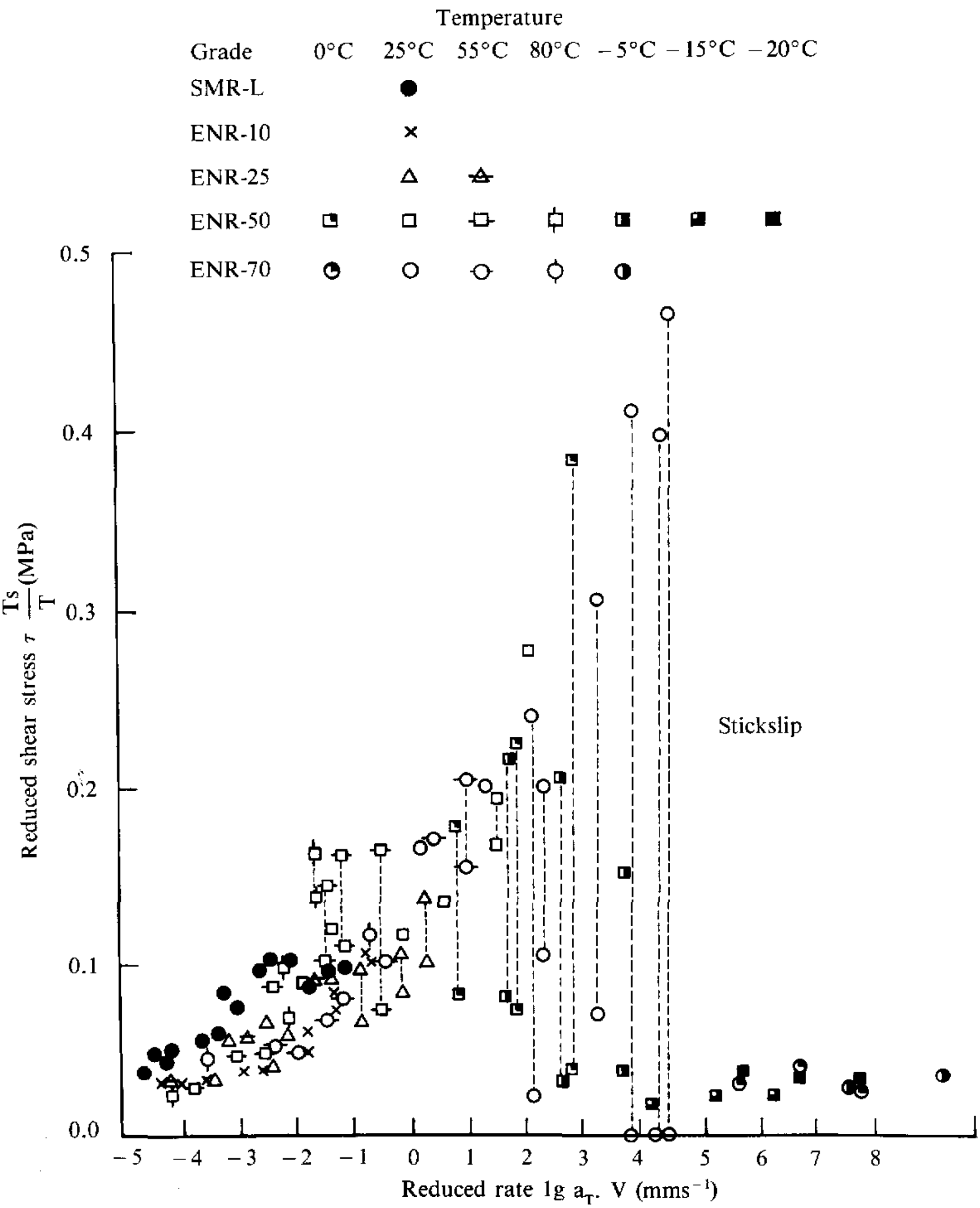

Figure 3. Effect of temperature and speed on interfacial shear stress for hemicylinders of SMR-L and different grades of ENR slid on a smooth steel track (0.06 μm CLA). Normal load 4.5N, RH = 55%–65%.

Influence of Surface Roughness

The effect of different degrees of track surface roughness on the friction coefficients of ENR-50 was investigated at a sliding speed of 1 mms^{-1}, using friction apparatus housed in a temperature cabinet[3]. This equipment allowed for more accurate force measurements and closer control over environmental conditions: the result was generally less scatter in the friction data. Roughening of a track was carried out with aluminium oxide and silicon carbide papers, and the resulting track roughness measured with a 'Talysurf' stylus profilometer. It was found that the friction was higher on a slightly roughened track when compared to smooth or very rough tracks *(Figure 4)*. This is similar in trend to other observations[2,8]. Measurements carried out at elevated temperatures of 55°C and 80°C also showed a similar increase in friction level on a slightly rough surface *(Figure 4)*.

It is noted that the smoothest surface used to obtain friction/roughness data *(Figure 4)* was chrome-plated steel, its measured roughness being 0.02 μm CLA. All other surfaces were roughened mild steel. A supplementary experi-

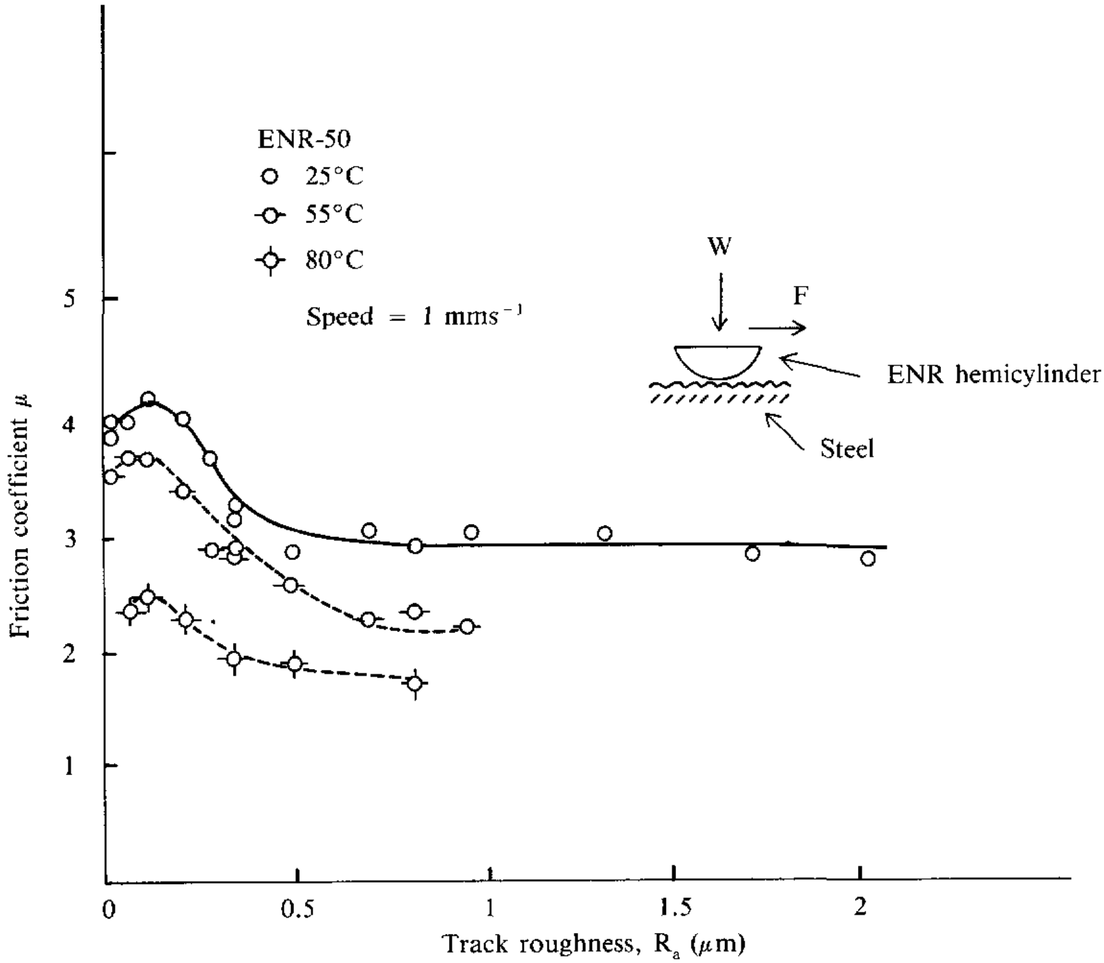

Figure 4. Friction of ENR-50 hemicylinders on steel tracks of different surface roughness maintained at different temperatures. Speed 1 mms^{-1}, normal load 4.5N, RH = 55%–65%.

ment was carried out in which an ENR-50 hemicylinder was drawn over silicon carbide paper that had an approximate roughness of 30 μm. It was interesting to find that the friction coefficient on this paper was $\mu = 3$, in line with the values at roughness of 1–2 μm.

The hemicylinders were examined visually after sliding against a roughened surface. At low roughness (less than 0.5 μm) a rubber ridge was always formed on the hemicylinder. With increasing roughness there was less rubber deformation into a ridge because the extended real area of contact was broken up into a series of point contacts. This led to 'pitted' abrasion patterns and no ridge formation at the higher roughnesses investigated. The patterns appeared to be formed by a combination of tearing[4] and yielding.

ENR Blends

Suitable amounts of unmasticated ENR-50 and SMR-L were mixed together on a two-roll mill to produce blends of Mooney viscosity ≈ 60 containing 25% and 10% levels of epoxidation, and then the friction values of these blends were measured. It was found that the friction levels of blends were comparable to that of ENR containing the same amount of epoxidation *(Table 1)*. Although only speculative, it was noticed that by using the empirical equation:

$$\mu_{\text{blend}} = \mu_A n_A + \mu_B n_B$$

where μ_A and μ_B are the friction coefficients

n_A, n_B are the mass fractions of the unmasticated component materials,

a reasonable estimate of the blend friction could be made *(Table 1)*. However, the blend friction may also be influenced by factors such as the extent of mastication and the stability of the component materials during blending. Also, the scatter in the friction data means some uncertainty in the argument for this simple law of mixtures, but it is offered as a thinking point.

Plasticisers as Surface Lubricants

Two commercial grades of plasticisers, namely Struktol A-60 and Struktol WB-16 (undisclosed mixtures of metal soaps of high molecular weight fatty acids, made by Schill and Seilacher) were evaluated for their influence on the surface friction of epoxidised natural rubber. They were cast from a volatile solvent onto the metal track. Typically it was observed that a 20%–25% reduction in friction level for rubber/metal contacts was achieved at room temperature. Lubrication was more effective at temperatures approaching the melting range of the plasticisers, the reduction in friction level being about 70% at 80°C *(Table 2)*.

TABLE 1. FRICTION OF ENR AND ITS BLEND WITH SMR-L

Grade/Blend	Friction, μ at different sliding speeds, V		
	0.2 mms^{-1}	0.7 mms^{-1}	1.3 mms^{-1}
ENR-25	2.1	2.6	2.9
ENR-50/SMR-L (1/1)	2.3	2.6	2.8
1/1 Blend estimate	2.3	2.6	3.2
ENR-10	1.8	2.5	2.9
ENR-50/SMR-L (1/4)	2.1	2.6	2.8
(1/4) Blend estimate	2.3	2.6	3.0
SMR-L (unmasticated)	2.3	2.7	2.9
ENR-50 (unmasticated)	2.2	2.6	3.5

Rubber hemispheres sliding on metal track under 4.5N load. Temperature 23°C–25°C, RH = 55%–65%. Friction quoted were average values; the scatter was about 20%.

TABLE 2. EFFECT OF PLASTICISERS ON ENR (BLENDED)

Plasticiser applied	Temperature (°C)	Friction coefficient	Observations
No plasticiser	25	2.6	Single ridge formed
	80	2.0	Transfer
Struktol A-60 film	25	2.0	Single shallow ridge
from toluene	80	1.1	Tearing and transfer, no plasticiser accumulation
Struktol WB-16 film	25	2.0	A thin layer of plasticiser film swept off by the specimen
(cast from ethanol)	80	0.9	
Mould release agent	25	<0.1	No apparent surface damage
(Addison Chemicals)	80	0.9	
Soap solution	25	0.3	No apparent surface damage
(10% Teepol)	80	0.1	

Sliding speed 0.2 mms^{-1}, load 4.5N, RH = 55%–65%. Average values of friction quoted, scatter 20%.
1/4 Blend ENR-50/SMR L.

When an excess amount of a mould release agent (Bomb-Lube, Addison Chemical) was sprayed onto the metal track at 25°C the sliding friction was reduced to almost zero. Partial evaporation of the volatile solvent in the mould released agent at 80°C led to an increase in the coefficient of friction to about 0.9.

Soap solution (10% Teepol) was found to be a very effective surface lubricant which reduced the coefficient of friction of a metal/ENR contact to 0.3 and 0.1 when measured at 25°C and 80°C respectively. Concomitant with this is a reduction in surface damage of the rubber specimen.

DISCUSSION

Both the temperature and level of epoxidation were found to influence the surface friction of raw ENR, but for any meaningful comparisons to be made it was necessary to take account of the sliding contact area as measured directly in the experiments. It then becomes possible to transform friction shear stress data into a single 'master-curve' by using the WLF equation. The added feature, compared to earlier published data[2] for SMR-CV, is that high values of the reduced rate parameter could be reached by using epoxidised rubber samples. The resulting master-curve resembles those[6,9] for vulcanised rubber. However, in making the comparison, certain differences are noted. For operating conditions adjacent to the glassy region very severe stickslip is encountered *(Figure 3,* centre); usually this arises if friction has a negative dependence on velocity, and in this case is presumably enhanced by the tackiness of the ENR. For conditions of high temperature/low speed (*Figure 3,* left hand side), the frictional stress is very low and becomes vanishingly small with decreasing rate. Here, presumably, the resistance to motion is mainly viscous. This behaviour may be contrasted with vulcanised rubber where it is believed that the more elastic-like material gives rise to a true static friction[10].

The enhanced friction at low surface roughness *(Figure 4)* is similar in trend to some observations[2,8] made with SMR-L and RSS 1, but the point to note, at least for ENR-50, is that the maximum largely occurs in the roughness range 0.1-1 μm CLA. This range is of technological interest. When processing raw rubber typical examples of surface finish encountered would be 0.8 μm CLA for the screw and barrel in an extruder, and 0.05–0.2 μm for CLA for well polished rotor surfaces in an internal mixer. The study here suggests that in this range the friction could nearly double with changes in surface roughness.

It is foreseen that there will be a need to blend ENR with NR for some applications, such as in tyre building, tyre tread formulations and engineering components. An understanding of the likely level of friction when processing such blends will be of benefit.

Process trials with ENR have shown that sticking problems can occur. For example, in mill mastication there appears to be safe temperature limits above which particular grades of ENR will start to stick onto the roller surface. The addition of certain accelerators to the raw rubber can ameliorate the problem. Plasticisers, such as zinc stearate, can also overcome the problem and hence a study of their lubricity for raw rubber/metal substrate contacts is of direct practical interest. Our studies *(Table 2)* clearly indicate that some materials are only effective at high temperatures, above their melting point, whereas others depend upon a different mechanism and become poorer lubricants at high temperatures.

CONCLUSION

The friction of raw ENR grades broadly varies according to glass transition temperature and, in common with other rubbers, according to ambient temperature and surface sliding speed. Data for various grades can be transformed into a single rate-temperature 'master-curve'. At low rates the frictional behaviour is akin to interfacial viscous shear with no true static friction, at intermediate rates stickslip motion tends to be present and at high rates 'glassy polymer' type friction ensues. Against slightly rough steel the friction can be greater than for a bright smooth finish or a grossly rough one. A simple prediction of the friction of NR/ENR blends seems possible for practical purposes and there is evidence for the action of plasticisers as surface lubricants.

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