J. nat. Rubb. Res., 5(1), 41-57

Effect of Process Aids on Surface Friction of Raw Rubber

E.L. ONG* AND A.D. ROBERTS**

The efficacy of various commercially available process aids as surface lubricants of raw natural rubber was examined using a simple friction apparatus. The aids were either spread on the hard counterface or incorporated in the raw rubber. Experiments were carried out over a range of temperatures from 25°C to 120°C at modest sliding speeds in order to follow changes in friction levels, surface morphology of the rubber and film transfer. In general, the presence of any process aid at the sliding interface led to a reduction in the surface friction.

Over the past decade or so, the rubber industry has become increasingly interested in the automation of machinery, and to accomplish this it requires rubber grades with predictable processing characteristics. To this end, process aids have a role to play. They are materials which can be added in small amounts to a rubber compound to improve processability in factory operations prior to vulcanisation, without adversely affecting the physical properties of the vulcanisate. For example, materials are available to provide lubrication so as to aid extrusion and moulding. Fatty acids and their derivatives, such as esters and metal soaps, increasingly find use as suitable materials. How such aids bring about processing improvements is not always clear, although there is evidence that they can lubricate molecules within the rubbery bulk. For example, measurements of stress relaxation in tensile testpieces of raw rubber indicate faster relaxation when an aid such as Dutrex 729 or Struktol A60, is incorporated into the rubber[1]. This can only be 'internal' lubrication.

Different process aids may act differently, perhaps depending on circumstances. They may be both internal and external lubricants. In mixing, it may be a balance between sufficient external friction and sufficient internal lubrication that is important and calls for an appropriate process aid. In die extrusion, however, the action of the aid may be essentially a surface effect to provide external lubrication[2].

A study of the surface friction of polypropylene showed that additives can have a profound effect[3]. As originally made, the polymer had high friction, but after storage the friction decreased. Reversion to the original friction could be obtained by washing the sample, indicating that the low friction was due to surface bloom of the additive. Such lubricants can be deliberately added to a polymer, or may be purely adventitious. The effect can be marked with only small quantities of additive, at the 0.1% level in the case of some surfactants.

A rheometer study[4] of the flow of unvulcanised rubber has shown that high levels of incorporated stearic acid reduce the friction at rubber-metal interfaces. It was inferred that a low viscosity/low shear strength phase separates out onto the rubber surface. The phenomenon is referred to as lubricated wall slip. Rubber compound formulations which retain lubricated wall slip at high shear rates do exhibit extremely smooth surfaces when extruded. Commercial stearic acid is a mixture of fatty acids with a range of carbon atom numbers. The rheometer study[4] was extended to examine the effect of fatty acid molecular weight on wall slip and it was found that the higher the molecular weight, the greater was the

*Rubber Research Institute of Malaysia, P.O. Box 10150, 50908 Kuala Lumpur, Malaysia
**Malaysian Rubber Producers' Research Association, Brickendonbury, Hertford SG13 8NL, United Kingdom

wall slip. It appeared that the lower molecular weight fatty acids less readily separated out to the surface. These results have control implications for factory operations.

This paper reports experiments on the lubricating effect of some commercial process aids by making friction measurements of unvulcanised Malaysian natural rubber sliding against a hard substrate with either an aid incorporated into the rubber or precipitated as a thin film on the substrate. In addition, tests are reported on using palm oil as an aid. Reduction in friction levels of up to 70% was found with certain types of process aid.

EXPERIMENTAL

Details of the process aids used in this study are given in *Table 1*. The raw natural rubber grades employed were SMR CV and SMR L. The formulations of the commercial process aids are not disclosed but they are believed to consist of mixtures of esters or metal soaps of long-chain fatty acids with melting points ranging from 50°C to 100°C. The mixing of any aid into a rubber was done on a laboratory two-roll mill (Brabender preparative mill). The roll surface was maintained at around 90°C. The addition of process aids (usually 5 p.p.h.r.) was carried out after the rubber had formed a continuous band on the mill surface, and the mixing process was continued for another 5 min.

Friction measurements were taken as an indication of the effectiveness of a particular process aid to act as a surface lubricant. The measurements were made with a simple apparatus[5]. A raw rubber hemisphere or hemicylinder was slid against a flat substrate of either 'Perspex' (polymethylmethacrylate), mild steel or a thin sheet of raw rubber supported on a hard substrate. Rubber hemicylinders and hemispheres were made by compression moulding at elevated temperatures[5]. When a 'Perspex' plate was used the sliding interface could be examined directly, and when

TABLE 1. TYPES OF PROCESS AIDS AND OTHER LUBRICANTS

Process aid	Chemical description	Melting point (°C)	Manufacturer
A	Esters mixture on inert carrier	50	S & S
B	Mixture of high molecular weight fatty acid derivatives	—	S & S
C	Water in oil emulsion	—	Kettlitz
D	Peptiser, water in oil emulsion	—	Kettlitz
E	Disperator	—	Kettlitz
F	Mixture of fatty acid soaps, mainly of calcium	95	S & S
G	Zinc soap of high molecular weight fatty acid	—	S & S
H	Zinc soap of high molecular weight fatty acid	—	Bozzetto
I	Zinc soap based on long- and branch-chain fatty acids	75–85	S & S

S & S = Schill and Seilacher

Many melting points cannot be quoted because the commercial process aid is carried on silica or other materials.

a mild steel plate was used its temperature could be raised to 120°C by an electric heater. A process aid was either put on a substrate surface as a thin film (spread, or precipitated from a solvent), or mixed into a rubber. Temperature measurements were made with a thermocouple probe touched onto a track surface. A sufficiently long contact dwell-time was allowed for the rubber sub-surface to warm up[5].

Under a normal load, W, the sliding friction force, F, was measured at a given speed and the friction coefficient, $\mu = F/W$, used to compare the efficacy of various process aids as surface lubricants of raw rubber. Unless otherwise stated, hard substrate surfaces were cleaned by wiping with tissue paper moistened with pure isopropanol, then allowed to dry. All rubber surfaces were tested as moulded without solvent treatment. The friction apparatus arrangement for measurements on a hot steel surface is shown in *Figure 1.*

Capillary Rheometry

Supplementary studies to examine the lubricity of process aids were carried out using a capillary rheometer (Monsanto Processability Tester) over a range of temperature and extrusion speeds[2]. The die capillary diameter was 2 mm and its length/diameter ratio was 20. This tends to give steady extrusion flow.

RESULTS

Sliding on 'Perspex' Track

A number of commercially available process aids were tested at room temperature for their lubricant action at the sliding interface of a raw SMR CV hemisphere (radius 21 mm) in contact with a length of 'Perspex' plate (polymethylmethacrylate), the track surface being smooth (surface roughness less than 0.1 μm Centre Line Average). A film of an aid was cast onto the 'Perspex' track. The various aids were compared for a fixed normal load at constant sliding speed. In general, the results *(Table 2)* show that all the process aids tested reduced the friction of a dry, clean track. The presence of water led to the lowest values of friction, and the least deformation or damage of the rubber surface.

Smooth-surfaced 10 mm thick sheets of raw SMR CV were laid on a 'Perspex' plate to form rubber tracks. Various aids were cast in turn

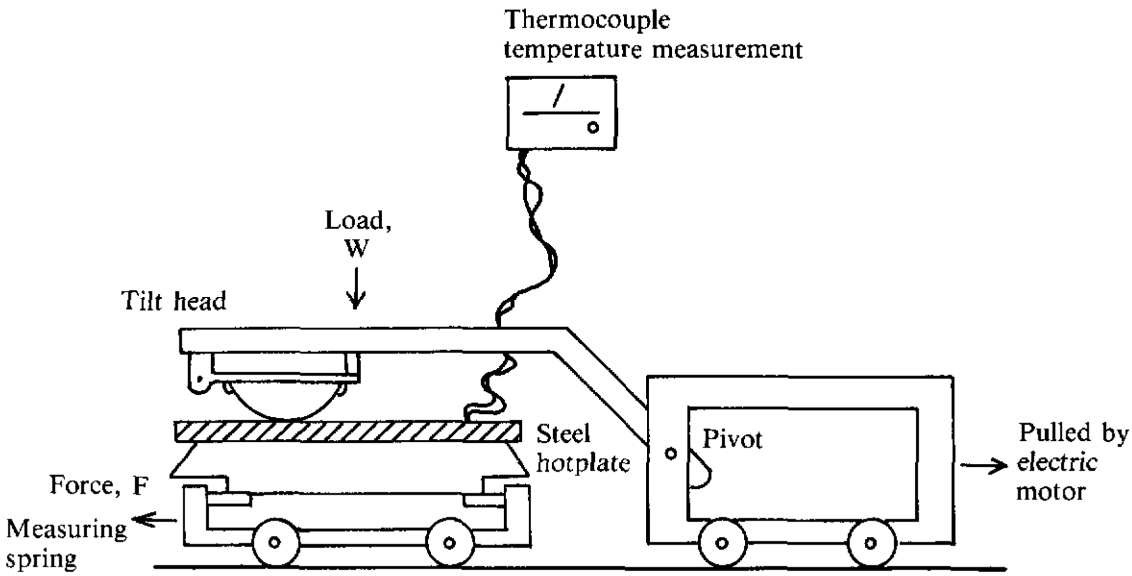

Figure 1. Apparatus for friction measurements against hot steel.

TABLE 2. EFFECT OF PROCESS AIDS ON SURFACE FRICTION OF SMR CV ON PERSPEX TRACK

Process aid	Track treatment	Friction coefficient	Observations on rubber surface
None	Dry	3.4	Ridge formation
A	Cast film	2.1	Some damage to the rubber
	Repeat cast	2.0	
	+ dist. H_2O	1.9	No damage
B	Cast, recast	2.0	Slight damage
	+ dist. H_2O	1.7	No damage
C	Cast, recast	1.7	Tiny ridges
	+ dist. H_2O	1.5	No damage
D	Cast film	2.1	Some scuffing
	Repeat cast	1.6	Slight scuffing
	+ dist. H_2O	1.4 ave.	No damage
	Cast film	1.1	No damage apparent
	Repeat cast	0.9	
	+ dist. H_2O	0.9	

Normal load 3.43 N; sliding speed 0.2 mms^{-1}, T = 22°C–24°C SMR CV hemisphere

onto the fresh tracks. Their lubricant effects, at room temperature, were compared when a hemisphere (radius 21 mm) of raw SMR CV was drawn over the track. All the process aids reduced the friction of untreated surfaces, some more than others *(Table 3)*.

Sliding on Mild Steel

Various process aids were tested by either putting them, in turn, on the mild steel track surface (surface roughness 0.2 μm CLA) or incorporating them into the raw rubber

TABLE 3. EFFECT OF PROCESS AIDS ON SURFACE FRICTION OF SMR CV HEMISPHERE ON SMR CV TRACK

Process aid	Track treatment	Friction coefficient	Observations on rubber surface
None	Dry	10	Severe surface damage
A	Cast film	5.1	Some slight damage
B	Cast film	9.4	Considerable damage
F	Cast film	6.3	Pulled out area of rubber
E	Cast film	3.2	Ridges on hemisphere, track damaged
D	Cast film	4.6	Ridges on hemisphere, track marks
C	Cast film	5.2	Ridges on hemisphere, track marks
Zinc stearate film		3.7	Ridges on hemisphere, track marks
G		2.1	Only slight surface damage
H		3.4	Slight surface damage

Normal load 3.43 N; sliding speed 0.2 mms^{-1}, T = 22°C–24°C.

(SMR CV). The rubber samples were in the form of moulded smooth-surfaced hemicylinders, (radius 21 mm). Friction measurements were carried out at room temperature and at selected elevated temperatures. The results *(Table 4)* indicate that at room temperature the reduction in the friction due to a process aid was generally greater when the aid was put on the steel track, than when it was incorporated into the rubber. With increasing temperature, the trend was for the difference in friction between surface deposition and rubber incorporation of an aid to become less. At higher temperatures, above the approximate melting point for the aids, surface or incorporated aid gave the same surface lubricating effect, absolute values of friction being noticeably less than at room temperature

TABLE 4. FRICTION ON MILD STEEL IN PRESENCE OF PROCESS AIDS

Process aid	Temperature of track (°C)	Friction coefficient		Friction stress (KPa)	
		For surface deposit	For rubber incorporation	For surface deposit	For rubber incorporation
F	24	0.7	2.4	28.0	77.4
	40	0.6	3.8	20.1	76.6
	60	0.5	2.0	10.5	18.3
	90	0.3	0.8	5.5	2.6
	120	0.2	0.4	2.5	1.2
G	24	1.7	2.1	74.7	53.1
	40	1.1	4.5	28.9	20.2
	60	1.5	2.0	25.4	5.4
	90	0.7	0.5	8.6	1.0
	120	0.8	0.3	7.1	0.5
I	24	1.9	2.0	55.6	49.2
	40	1.7	3.8	43.6	20.1
	60	0.7	1.3	12.3	3.4
	90	0.5	0.7	6.2	1.0
	120	0.4	0.3	3.7	0.6
H	24	1.2	2.1	51.6	85.4
	40	1.2	3.9	35.4	25.2
	60	1.6	1.6	23.0	5.2
	90	0.3	1.3	5.2	3.1
	120	0.2	0.4	2.5	0.9
No aid	24	3.7		392	
	40	3.5		261	
	60	2.8		133	
	90	1.8		52.1	
	120	1.2		13.6	

Normal load 3.43N; sliding speed 0.2 mms^{-1}

(Figure 2). With increasing temperature, surface damage due to sliding of rubber samples became less.

A peak in the friction coefficient that occurred at 40°C was observed for all aids when they were incorporated into rubber *(Table 4, Figure 2).* This appears to be due to a softening effect. When an aid was incorporated into a rubber sample, the body of the sample readily deformed upon loading against the steel track, with the result that the real contact area was much enlarged compared to a similar sample without aid incorporated. This enlarged contact area is believed to be the reason for the peak friction at 40°C. An examination *(Figure 3)* of contact areas after sliding was found helpful to interpretation. A plot of the friction stress (friction force/contact area) shows the efficacy of a process aid above its melting temperature *(Figure 4).* When allowance is made for the actual contact area a 'cross-over' in friction levels is found, the lower level at high temperature resulting from process aid incorporated into the rubber. One might surmise that the cross-over in frictional stress levels above 80°C could be due to the aid-incorporated rubber having a softer bulk. For a given load, this would lead to a relatively higher real area of contact and hence high friction. This result, obtained in the laboratory for a rubber hemisphere slid against a steel plate, may not be borne out in practical situations of different contact geometry.

Capillary Rheometry

The aim of these rheometry studies was to see the amount by which the apparent capillary shear stress is reduced at a given shear rate by the incorporation of different process aids. In an earlier study[2] of flow curves, it was found that the viscosity was reduced by the addition of a particular process aid. The results *(Table 5)* of this study show that an incorporated process aid can reduce the shear stress considerably, particularly at high temperature and low shear rate under conditions of steady extrusion. For

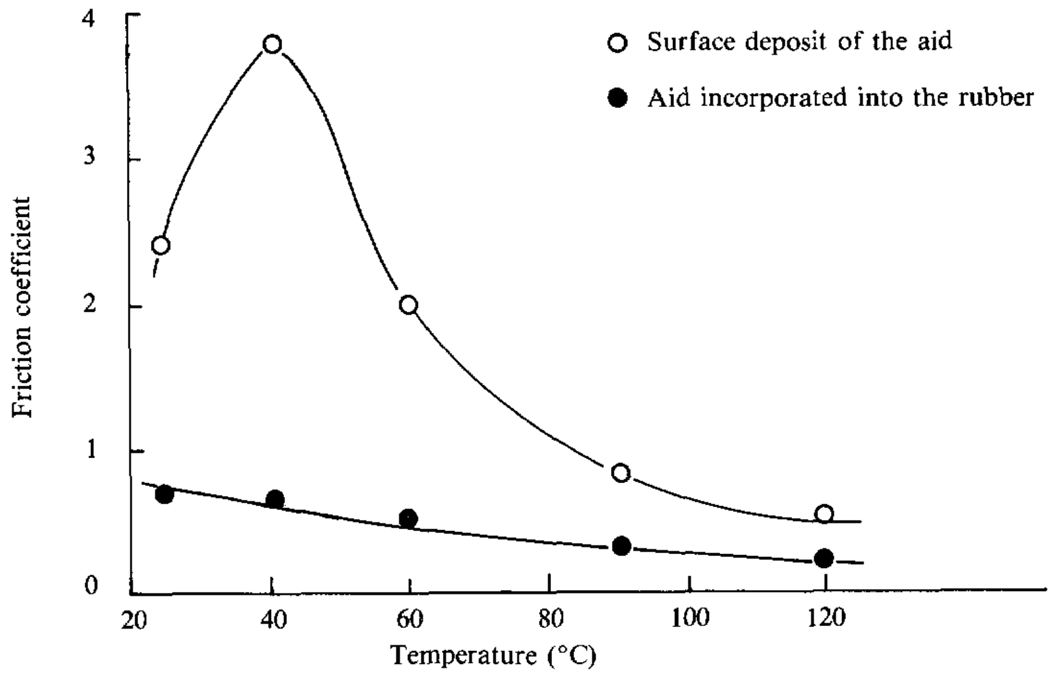

Figure 2. Effect of process aid F on friction of SMR CV against steel.

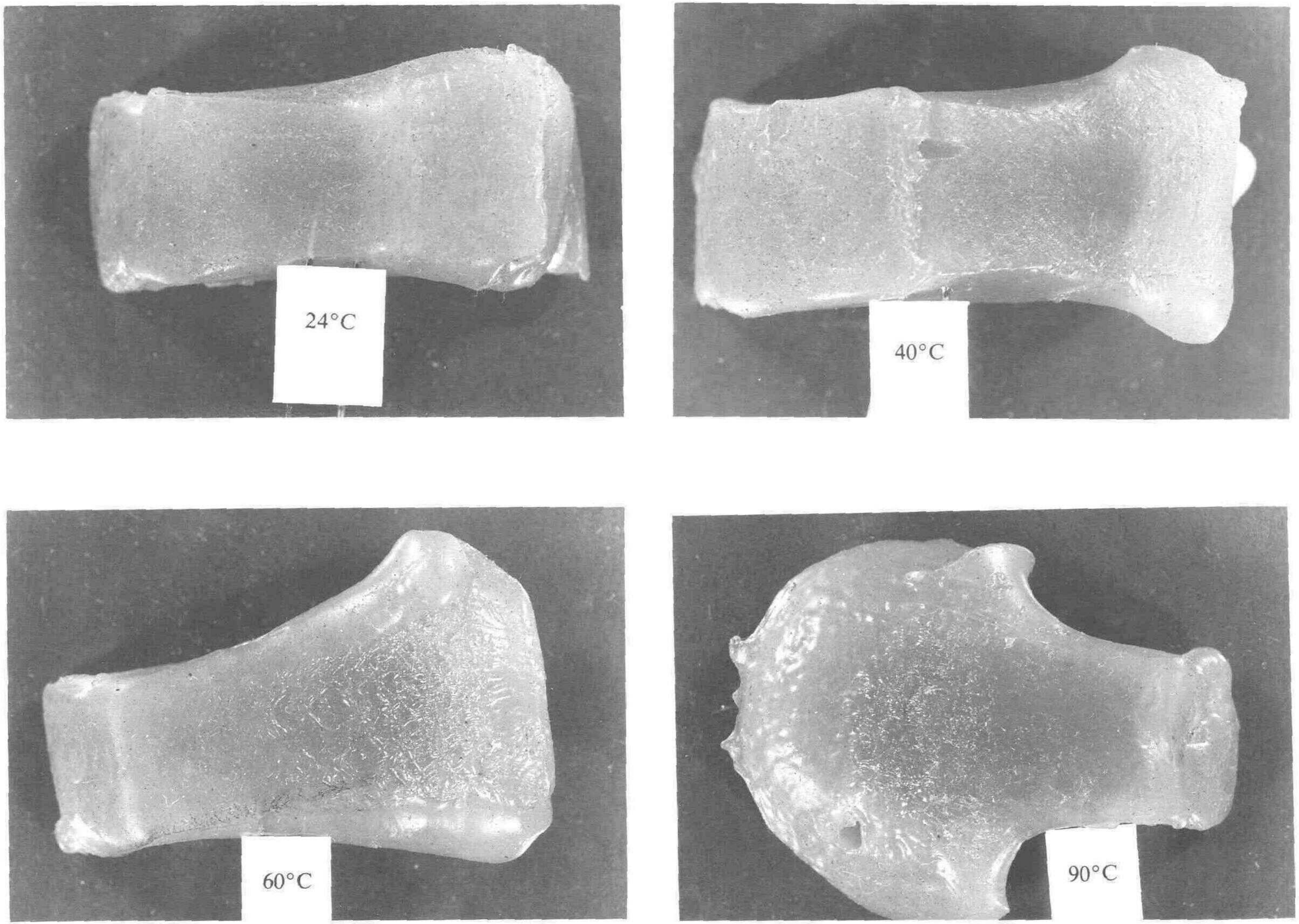

Figure 3. Contact areas as seen after sliding of SMR CV against steel. The rubber contained the process aid F incorporated into its bulk. Steel surface temperatures were 24°C, 40°C, 60°C and 90°C.

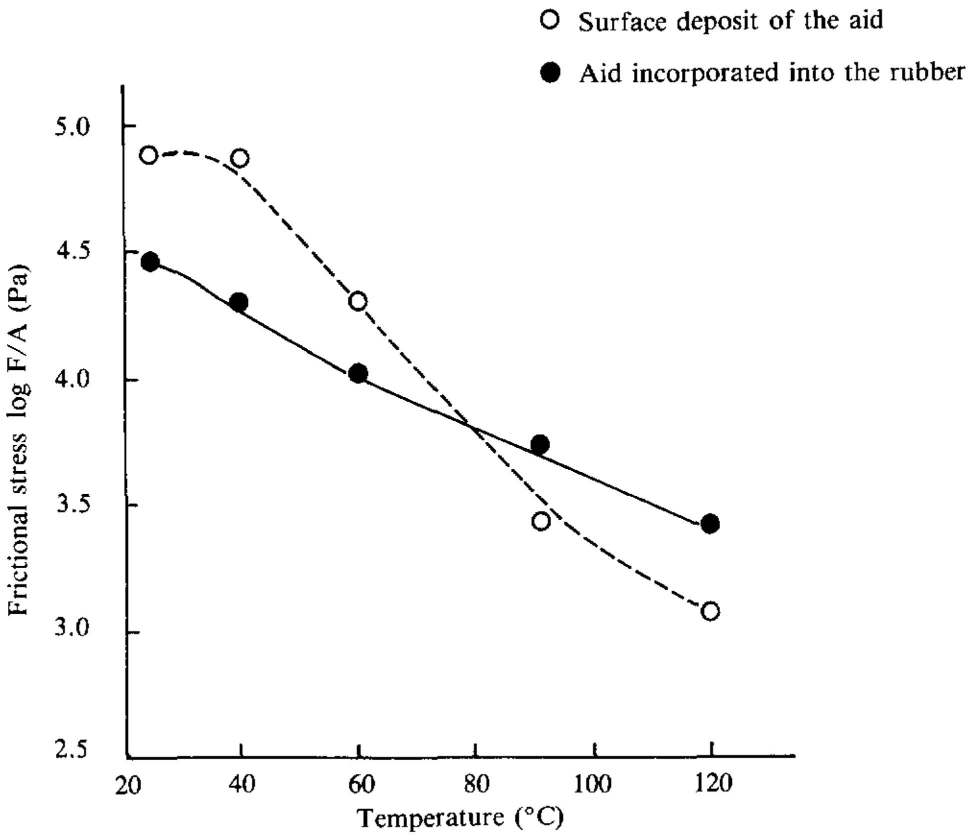

Figure 4. Plot of friction stress to show efficacy of the process aid F above its melting point.

TABLE 5. EXTRUSION STRESS AT DIFFERENT APPARENT SHEAR RATES OF SMR CV WITH VARIOUS PROCESS AIDS INCORPORATED

Process aid	Temperature (°C)	Extrusion stress (MPa)			
		1.5 s^{-1}	7.6 s^{-1}	15.1 s^{-1}	76 s^{-1}
I	60	0.072	0.127	0.149	0.243
	90	0.039	0.071	0.086	0.143
	120	0.017	0.042	0.057	0.101
G	60	0.075	0.140	0.172	0.303
	90	0.034	0.061	0.084	0.144
	120	0.012	0.026	0.039	0.084
NR (control)	60	0.269	0.582	0.765	1.045
	90	0.140	0.194	0.216	0.280
	120	0.107	0.162	0.194	0.248

Conditions: L/D = 20, diameter 2 mm

these circumstances, the measured friction stress for incorporated process aid *(Table 4)* mirrors this behaviour.

Palm Oil

The objective was to discover whether palm oil might provide effective surface lubrication of raw natural rubber, and whether it also plasticises the rubber. Accordingly, testing of SMR L hemicylinders sliding against mild steel was done. Palm oil in various states of refinement was either deposited onto the steel surface, or it was incorporated into the rubber bulk. Friction measurements were obtained at different temperatures, and a comparison made with epoxidised soya bean oil which is used industrially to plasticise various plastics such as PVC. The results *(Table 6)* suggest that crude palm oil acts as a good lubricant when incorporated into the rubber. Refined states or components do not show any improvement in lubricity. The surface deposit was particularly effective because a lot of oil was used. It seems surprising that rubber incorporated soya bean oil which is less compatible with rubber than palm oil gives higher friction values at high temperatures *(Table 6)*. It is possible that there

TABLE 6. FRICTION COEFFICIENTS FOR PALM OIL AS A PROCESS AID FOR SMR L

Oil	Temperature (°C)	Friction coefficient	
		Surface deposit	Rubber incorporation
Zero oil	24		4.1
Epoxidised crude palm oil	24	0.22	1.5
	40		0.8
	60		0.3
	90		0.3
Epoxidised RDB palm oil	24	0.09	1.5
	40		0.4
	60		0.6
	90		0.4
Epoxidised RDB palm stearin	24	0.11	1.5
	40		0.7
	60		0.6
	90		0.3
Epoxidised RDB palm olein	24	< 0.01	1.6
	40		0.7
	60		0.3
	90		0.3
Soya bean oil (epoxidised)	24	0.11	1.5
	40		0.5
	60		0.5
	90		0.5

RDB — Refined de-odourised bleached   Sliding speed 0.2 mms^{-1}

TABLE 7. EXTRUSION STRESS OF SMR L WITH VARIOUS OILS INCORPORATED AT VARIOUS APPARENT/SHEAR RATES

Oil	Extrusion stress (MPa)							
	$3.6\ s^{-1}$	$10.8\ s^{-1}$	$21.6\ s^{-1}$	$36.0\ s^{-1}$	$108.0\ s^{-1}$	$218.0\ s^{-1}$	$360.0\ s^{-1}$	$720.0\ s^{-1}$
None (control)	0.237	0.284	0.315	0.345	0.491	0.717	0.872	1.138
Crude palm oil	0.197	0.235	0.257	0.278	0.345	0.483	0.707	1.000
Epoxidised crude palm oil	0.198	0.234	0.252	0.269	0.322	0.427	0.528	0.976
RBD palm olein	0.200	0.233	0.248	0.265	0.315	0.425	0.500	0.961
Epoxidised RBD palm oil	0.198	0.237	0.259	0.276	0.339	0.465	0.612	0.974
RBD palm oil	0.190	0.222	0.240	0.254	0.303	0.384	0.481	0.871
Epoxidised RBD palm olein	0.200	0.248	0.267	0.284	0.338	0.447	0.690	1.045

RBD = Refined de-odourised bleached
Extrusion at 100°C

is a smaller amount of soya bean oil being incorporated at the mix stage of the rubber to migrate to the surface to lower the friction. Its compatibility may also change with temperature. This will be studied further.

The capillary rheometer (Monsanto Processability Tester) was employed to examine the effect of palm oil on extrusion flow. The results *(Table 7)* show that crude palm oil can reduce extrusion stresses but no further reductions result from refinement of the oil. Thus the friction and extrusion data indicate that crude palm oil is quite sufficient as a process aid, without any refinement.

Effect of Presence of Water

From time to time, there are reports that the presence of 'knuckles' or wet spots in raw rubber can give rise to processing problems due to slip during mixing. This can affect the dispersion of formulation ingredients. A simple study shows *(Table 8)* by how much the friction might change. Knuckles were deliberately introduced by dipping the surface of rubber hemicylinders in distilled water for 72 h. The surface was then wiped dry before friction experiments were conducted.

DISCUSSION

Whatever the operating temperature, the presence of a process aid at the sliding interface leads to a reduction in friction. However, it is apparent that such materials are most effective above their melting point. For example, at room temperature, the majority of the process aids studied could about half the friction, but at 120°C, the friction typically fell to a fifth of the value in the absence of an aid at the same temperature. A comparison of frictional stresses further shows that at high temperatures, an aid is more effective when incorporated into the rubber.

Water lubrication by 'knuckles' reduces the friction at room temperature by a factor of two, but at elevated temperature (90°C), there is only a 20% difference. This lack of lubrication at high temperatures may be contrasted with the behaviour of process aids.

Palm oil appears capable of providing reasonable surface lubrication when incor-

TABLE 8. EFFECT OF 'KNUCKLES' ON FRICTION

Temperature (°C)	Knuckles in rubber	Friction coefficient
25	None	4.1
90	None	2.0
25	With knuckles	1.9
90	With knuckles	1.5

Mild steel track, sliding speed 0.2 mms^{-1}, normal load 3.43 N porated into raw rubber. No advantage is gained from refining it. Whether it can be considered a viable alternative to currently used process aids may depend upon precise operating circumstances, but at least it is more compatible with rubber than some other oils. The effects of palm oil and its derivatives on the physical properties of rubber vulcanisates need to be studied before definitive recommendations can be made. It is interesting to note that some of the commercially available process aids are in fact derived from palm oil.

CONCLUSION

It is found that commercial process aids are most efficient above their melting point when they form a lubricating film at the rubber interface. This may result in a fall in friction by up to six times. Crude palm oil can act as a process aid and might merit further consideration for industrial use.

ACKNOWLEDGEMENT

The authors thank Encik Abdul Majid bin Osman for his technical assistance and Dr Hamirin Kifli of Palm Oil Research Institute of Malaysia for supplying the palm oil and its derivatives.

Date of receipt: August 1989
Date of acceptance: December 1989

REFERENCES

1. FULLER, K.N.G. Unpublished Data. Malaysian Rubber Producers' Research Association, United Kingdom.
2. ONG, E.L. AND LIM, C.L. (1985) Effect of Process Aids on the Processing Behaviour of Natural Rubber. *Proc. Rubb. Conf. 1985 Kuala Lumpur*, **2**, 622.
3. HUXTABLE, J., COGSWELL, F.N. AND WRIGGLES, J.D. (1981) Polymer-metal Friction as Relevant to Processing. *Plastics and Rubber Processing and Applications*, **1**, 87.
4. TURNER, D.M. (1984) The Contribution of Friction to the Flow of Unvulcanized Rubber. *Institute of Physics Tribology Group Meeting, Hertford, United Kingdom.*
5. ONG, E.L. AND ROBERTS, A.D., (1983) Experiments on the Friction of Raw Natural Rubber. *J. Rubb. Res. Inst. Malaysia*, **31(3)**, 236.