

Factors Affecting the Coefficient of Friction of Paper

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SYNOPSIS

Electron spectroscopy for chemical analysis (ESCA) studies on a variety of paper samples show that for samples of similar surface roughness the coefficient of friction is a function of the amount of extractives that are present on the sheet surface. Corona discharge treatment in air is found to produce a marked increase in the coefficient of friction of newsprint due to surface oxidation effects. © 1992 John Wiley & Sons, Inc.

INTRODUCTION

The friction properties of paper are important during finishing, converting, and end-use operations. In spite of this, it is surprising that very little is known of the factors affecting paper friction. Previous work in the field focuses on the effect of surface roughness and operating conditions¹⁻⁴; however, the conclusions drawn in these reports are often mixed and contradictory. For example, Broughton and Gregg report that when the sheet surface is smoothed by calendering the coefficient of dynamic friction (paper against metal) is reduced and conclude that friction is caused by surface asperities of the sheet.¹ On the other hand, Jones and Peel conclude that calendering has no effect on the coefficient of dynamic friction.³

In a review article, Tabor describes the friction behavior of polymers in terms of adhesion and deformation processes.⁵ It is found that the adhesion component of friction depends upon the viscoelastic properties of the polymer and that sliding speed, temperature, and contact pressure are important factors.

Another important aspect of polymer friction is boundary lubrication effects where thin surface films of aliphatic compounds such as fatty acids can act as lubricants at the polymer/polymer or polymer/metal interface, thus reducing friction forces.⁶

It is conceivable that for a cellulosic, viscoelastic material such as paper some of the factors governing polymer friction may also apply. For example, surface topography and conformability of the sheet surface under a given load will govern the contact area and likely affect the friction properties. In terms of boundary lubrication effects, there have been model experiments showing that deposition of long-chain fatty acids on paper surfaces can reduce the coefficient of friction.^{1,7,8}

In this study, we examine the friction properties of a range of newsprint paper samples under constant loading and sliding speed. It is assumed that the surface conformability of the samples is similar under the loading conditions that are used. The focus of the study is on the role of natural wood extractives on paper friction. Electron spectroscopy for chemical analysis (ESCA) is used to get an indication of the elemental composition of the paper surface and provide a means of estimating the extent of extractive deposition.

EXPERIMENTAL

The surface topography of the newsprint samples were characterized by determining the root mean square profilometric roughness with a Taylor-Hobson Model 3 profilometer. The stylus of the instrument was a four-sided 90° diamond pyramid with a rounded tip about 2.5 microns wide. The force of the stylus on the surface of the sheet was about 100 mg and the measurement was made over a distance of 2 mm. The Parker Print Surf (PPS) (S10)

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roughness was also measured on some of the samples. The profilometric and PPS roughness are reported as an average of 15 readings performed on different samples. Photomicrographs of the paper surfaces were also obtained under 15° light illumination along the paper machine direction to obtain a qualitative view of the surface texture.

The ESCA spectra were run by a commercial analysis service (Surface Science Laboratories, Mountain View, CA). Samples were prepared and handled with care by employing carefully washed surgical gloves to avoid contamination.

The coefficient of dynamic friction between a chrome-plated metal disc and the paper top surface was determined by sliding a 37-mm diameter disc weighing 103.9 g across the sheet with a frictionless pulley attachment that was attached to the load cell of an Instron tester. The crosshead speed was 50 cm/min. The results are the average of 10 readings performed on different samples. To ensure that there was no contamination of the samples, clean surgical gloves were worn and the metal disc was cleaned with acetone after each test.

The coefficient of static friction (paper against paper, top side/bottom side in the machine direction) was measured using two different methods. In the first method, the slide angle was measured using an inclined plane tester as recommended in the TAPPI standard (T-815). The coefficient of static friction was then calculated by taking the tangent of the slide angle. Five pairs of sheets were used and each pair was tested five times. The results that are reported are the average of 25 readings. In the second

method, the horizontal plane technique was used; the load cell of an Instron tester was employed to determine the force required to create the onset of sliding motion. Ten pairs of sheets were used and each pair was tested once.

The sheets were treated with the extractive components by dipping them into the appropriate solvent (chloroform or dichloromethane) containing different concentrations of the contaminant. The sheets were then aerated and reconditioned at 50% relative humidity and 23°C prior to testing.

The newsprint samples were treated in an air corona discharge at 12,000 V for periods ranging from 5–300 s. Both sides of the sample were treated in the corona unit. The equipment and detailed procedure are described in an article by Goring.⁹

RESULTS AND DISCUSSION

Effect of Surface Topography on Friction

Newsprint samples were obtained from various mills representing different processes and wood species. The coefficient of static friction was measured with an inclined plane tester; these are shown along with the PPS and the root mean square profilometric roughness values in Table I. Photomicrographs under 15° light illumination were obtained on all samples; some examples are shown in Figure 1. The results in Table I and Figure 1 indicate that for the range of roughness values encountered here there is no correlation between roughness and friction. For

Table I Coefficient of Static Friction of Newsprint Samples Obtained from Various Mills

Sample	Coefficient of Static Friction ^a	PPS (S10) Roughness (μm)		Average Profilometric Roughness (μm)	
		Top Side	Bottom Side	Top Side	Bottom Side
A	0.47	—	—	—	—
B	0.35	—	—	—	—
C	0.50	—	—	—	—
D	0.36	—	—	—	—
E	0.32	—	—	—	—
F	0.59	3.40	3.45	2.91	2.93
G	0.44	3.14	3.55	2.60	2.74
H	0.55	3.69	3.56	3.20	3.19
I	0.59	—	—	—	—
J	0.49	3.36	3.55	3.35	2.55

^a Paper against paper.

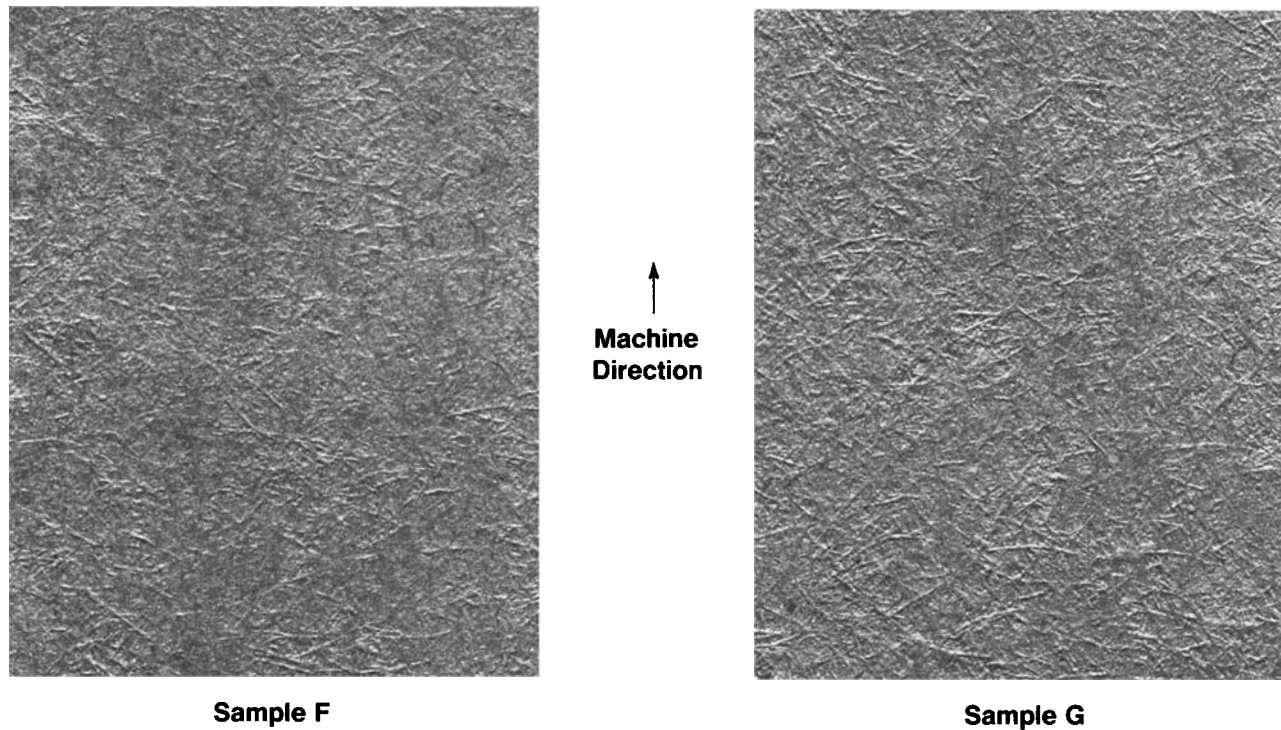


Figure 1 Surface topography of the top side of newsprint samples F and G photographed under 15° light illumination along the machine direction.

example, sample F has a higher coefficient of static friction (COF) value than sample G even though it appears to have smoother surface features (Fig. 1). Similar trends were observed by Back, who found no correlation between PPS roughness and static friction for samples ranging from newsprint to magazine paper.⁸ These results imply that other factors are responsible for the differences in friction value. Since previous work implied that surface lubrication effects may be important for paper friction, we decided to look in depth at this phenomena.

ESCA Analysis of Extractive Deposition on Paper Surfaces

Principles of the Method

ESCA gives an estimate of the atomic composition of the surface layer of solids and provides some indication of chemical bonding.¹⁰ The sample is irradiated with monochromatic X-rays; this in turn causes the emission of photoelectrons from the inner-shell orbitals of atoms in the sample. The photoelectrons emitted from the bulk of the sample lose their energies through repeated collisions. Thus, only atoms at a limited depth (5 nm) contribute to

the intensity of the measured electron emission, making it possible to analyze the surface layer of the sample. The energy of the emitted photoelectrons is proportional to the binding energy that held the electron to the parent atom. Since every element has a characteristic binding energy, the different atoms present on the surface may be identified easily.

The nature of the chemical bonds at the surface layer of a solid may be inferred from ESCA experiments. The energy of electrons emitted from a given elemental shell may be altered depending upon the type of chemical bond formed by the element in question. This “chemical shift” may indicate the type of chemical bonds present on the surface. Binding energies and chemical shifts for carbon (1s) atoms in organic compounds have been measured and tabulated. Based upon these data, the carbon atoms in wood components may be divided into four broad classes, in order of increasing chemical shifts^{11,12}:

- C₁: Carbon atoms bonded only to carbon and/or hydrogen.
- C₂: Carbon atoms bonded to a single oxygen other than a carbonyl oxygen.
- C₃: Carbon atoms bonded to two noncarbonyl oxygens or to a single carbonyl oxygen.

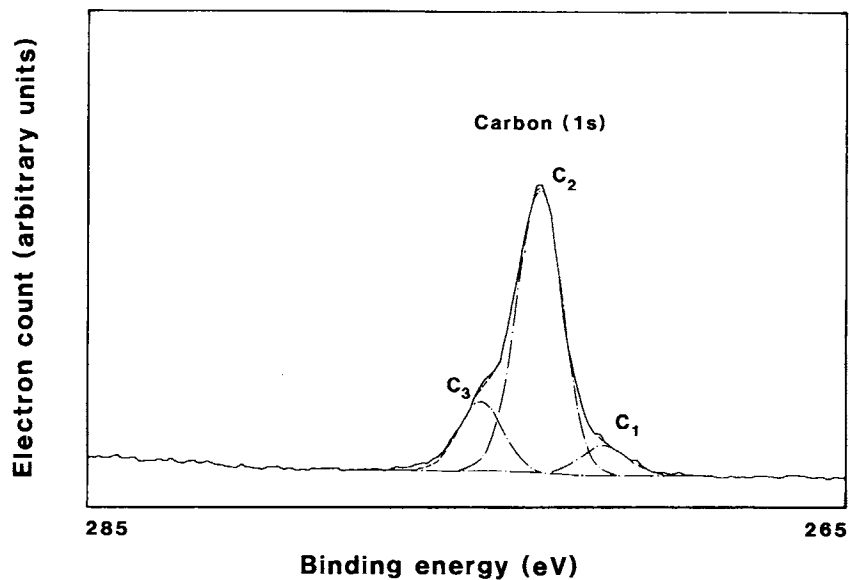


Figure 2 ESCA carbon (1s) spectrum of an extracted Whatman No. 1 filter paper sample.

C₄: Carbon atoms bonded to a carbonyl and a noncarbonyl oxygen.

The electron binding energies associated with each class increase from class C₁ to C₄.

Figures 2 and 3 illustrate the carbon and oxygen ESCA peaks for an extracted sample of Whatman #1 filter paper. This sample approximates to pure cellulose. The carbon peak has been resolved into

three components with the C₂ component being the dominant one for pure cellulose. The C₄ component was not detected. The area under the peaks corresponds to the number of carbon and oxygen atoms on the surface; thus, a ratio of oxygen atoms to carbon atoms (O/C) may be obtained. This ratio gives a direct measure of the hydrophobicity of the surface with a low O/C ratio signifying a hydrophobic sur-

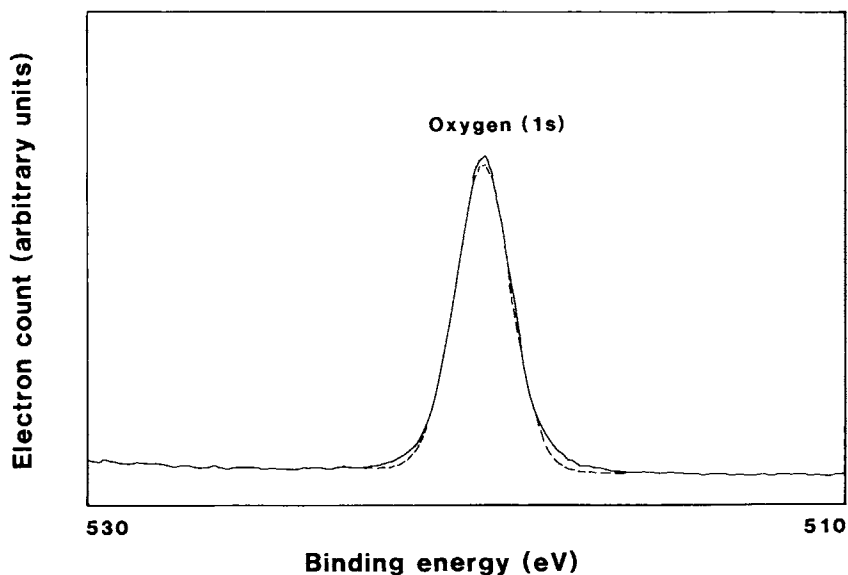


Figure 3 ESCA oxygen (1s) spectrum of an extracted Whatman No. 1 filter paper sample.

face. For a pure cellulose surface, the theoretical O/C ratio is that of the anhydroglucose unit, 0.83. The Whatman filter paper sample in this study gave a value of 0.73, which is somewhat lower than the theoretical value; this could be due to damage from the X-ray source of the spectrometer.¹³

Influence of Extractives on the Friction Properties of Paper

Wood extractives are highly surface active and are deposited on the surfaces of fibers. During chemical pulping, most of these extractives are solubilized and are washed out with the cooking liquor; this is not the case for mechanical pulps. Thus, newsprint furnishes are likely to contain extractives on their surface; due to their "slippery" nature, these hydrophobic materials can then act as lubricants, reducing the friction coefficient.^{1,7,8}

Preliminary experiments were conducted with pure cellulose filter paper to monitor the effect of extractives on the friction properties. Samples of paper were immersed in solutions of a common wood extractive, glycerol trioleate ($C_{57}H_{104}O_6$), in chloroform of varying concentration. The solvent was evaporated and the oxygen to carbon atom ratios were determined by ESCA. The coefficient of dynamic friction between a chrome-plated metal disc and the treated paper surface was measured with an Instron tester. The metal disc was cleaned after each test with acetone to prevent contamination of the samples. Table II shows that with the addition of extractives both the coefficient of friction and the O/C ratio decreased significantly, indicating that O/C ratio is related to friction. At the highest treatment level, the O/C ratio of 0.15 indicates that the cellulose surface is uniformly covered with the extractive (theoretical O/C value for total coverage is 0.10, 6 oxygen atoms to 57 carbon atoms). At lower coverages, the friction coefficient is also reduced, indicating that small quantities deposited on the surface can still have a significant effect. The effect of the hydrocarbon contaminant on the carbon peak shape is illustrated in Figures 4 and 5 for two levels of treatment; compared to a noncontaminated surface (Fig. 2), the C_1 peak intensity increases significantly, showing the abundance of extractives on the paper surface.

The newsprint samples listed in Table I were extracted for 4 h using high-purity grade chloroform in a soxhlet extractor. These samples along with the unextracted sheets were sent for ESCA analysis. Measurements were made on both top and bottom

Table II Effect of Extractives on the Coefficient of Dynamic Friction of Whatman No. 1 Filter Paper

Concentration of Glycerol Trioleate in Chloroform (mol/L)	Coefficient of Dynamic Friction ^a	Oxygen/Carbon Atom Ratio
0.000	0.40	0.73
0.002	0.30	—
0.003	0.26	0.33
0.006	0.28	—
0.057	0.21	—
0.090	0.17	0.15

^a Paper against metal.

sides of the samples. Survey scans were also made on the original samples; these scans cover a broad range in the binding energy, ranging from 0–1000 eV in search of different elements. On these samples, the only elements detected were carbon and oxygen. Table III is a summary of the O/C ratios and the coefficient of static friction between paper/paper surfaces measured on an inclined plane tester. Upon extraction, all samples have similar O/C ratios; thus, any differences in the O/C ratios of the unextracted samples are due to extractives content. Once again, a low friction value is associated with a decrease in the oxygen to carbon atom ratio. The carbon spectra of samples F and G are shown in Figures 6 and 7. The C_1 class of carbon atoms (271 eV binding energy) represents C—H or C—C bonds, which are associated with aliphatic type compounds such as long-chain fatty acids. Thus, sample G has more of these chemical components on its surface compared to sample F and as a result a lower friction coefficient. The ESCA analysis data shown in Table III were obtained from the top side of the samples; similar results were obtained on the bottom side of the samples.

Corona Treatment of Newsprint

Corona treatment in air or oxygen produces ozone gas, which can degrade and oxidize polymeric surfaces. The treatment is widely used to improve wettability and adhesion.¹⁴ The use of corona discharge on paper surfaces is less common; the first experiments on cellulose strips were carried out by Goring.⁹ In another study, the wettability of corrugating medium was enhanced through surface oxidation in a corona discharge.¹⁵

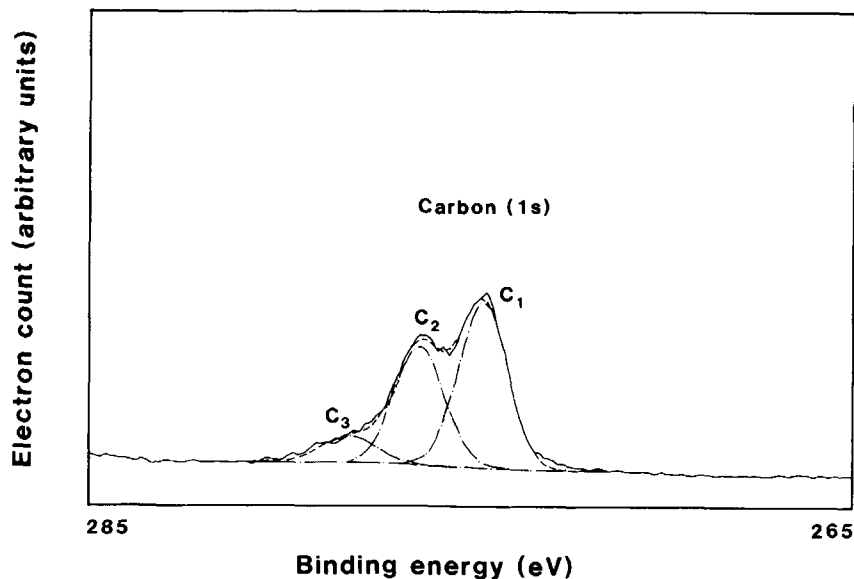


Figure 4 ESCA carbon (1s) spectrum of Whatman No. 1 filter paper coated with a solution of glycerol trioleate in chloroform (0.003 mol/L).

A treatment that oxidizes the paper surface and renders it hydrophilic is likely to affect the coefficient of friction. Recently, Back showed that corona treatment of kraft liners increases the coefficient of friction; however, the effect of the treatment was not permanent.⁸

A newsprint sample was treated in an air-corona discharge at 12,000 V for periods up to 300 s. A sam-

ple of the paper before and after the corona treatment was sent for ESCA analysis. Coefficient of static friction (paper against paper) was measured with an inclined plane tester. The results in Table IV indicate that corona treatment increases the coefficient of friction by 30% after 30 s of treatment; further treatment (300 s) did not seem to improve the effect. The friction values of the treated samples

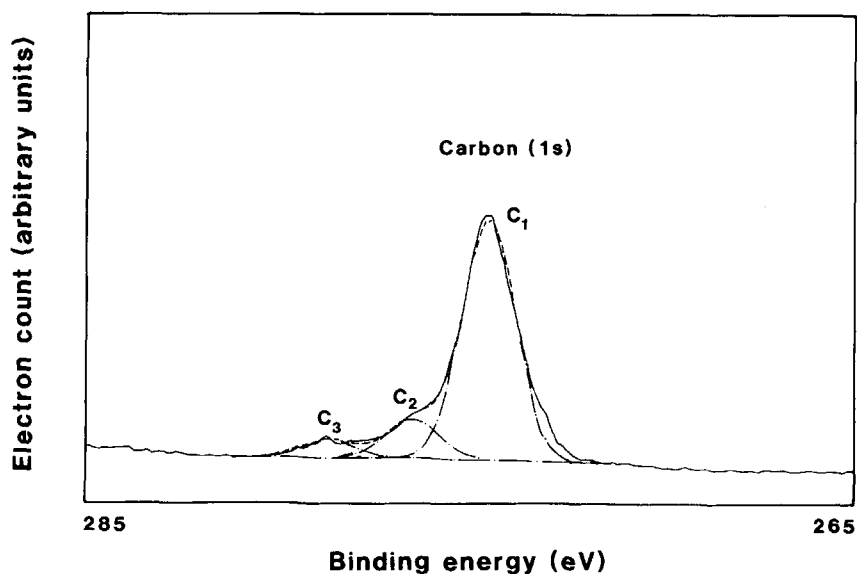


Figure 5 ESCA carbon (1s) spectrum of Whatman No. 1 filter paper coated with a solution of glycerol trioleate in chloroform (0.090 mol/L).

Table III Coefficient of Static Friction and Electron Spectroscopy Analysis of a Variety of Newsprint Samples

Sample	Coefficient of Static Friction ^a	Oxygen/Carbon Atom Ratio ^b	Oxygen/Carbon Atom Ratio ^b After Extraction
A	0.47	0.37	0.55
B	0.35	0.30	0.55
C	0.50	0.42	0.55
D	0.36	0.33	0.55
E	0.32	0.31	0.55
F	0.59	0.44	0.56
G	0.44	0.37	0.56
H	0.55	0.44	0.55
I	0.59	0.44	0.56
J	0.49	0.38	0.55

^a Paper against paper.

^b Measured on the top side of the sample.

were remeasured 8 days after the treatment and were found to be the same, indicating that the changes may be permanent. The oxygen-to-carbon-atom ratio increased from 0.39 to 0.47 after 300 s of treatment, indicating that oxidation of the molecular components on the sheet surface occurred. It is not clear why corona treatment of paper increases fric-

tion, but it is well known that for polymers improvement in adhesion occurs by incorporation of polar groups.¹⁴ A similar mechanism may be operating here.

Relationship between Friction and the Oxygen-to-Carbon-Atom Ratio

The data from Tables III and IV were used to generate a plot of the coefficient of static friction (paper against paper) as a function of the oxygen/carbon atom ratio (Fig. 8). The coefficient of determination, r^2 , was 0.90, indicating that the coefficient of static friction is highly correlated with the O/C atom ratio. This suggests that the static friction coefficient is a function of the degree of hydrophobicity of the paper surface. This arises from variation in surface extractives content (samples from Table III) or the degree of oxidation of the paper surface (samples from Table IV). The variation in the surface extractives content is likely to be a function of different wood species used and the various processing conditions.

Effect of Type of Extractive Material

Extractives are a heterogenous mixture of materials ranging from relatively low-molecular-weight fatty acids, resin acids, and sterols to the higher-molecular-weight steryl esters and glycerides. Physically,

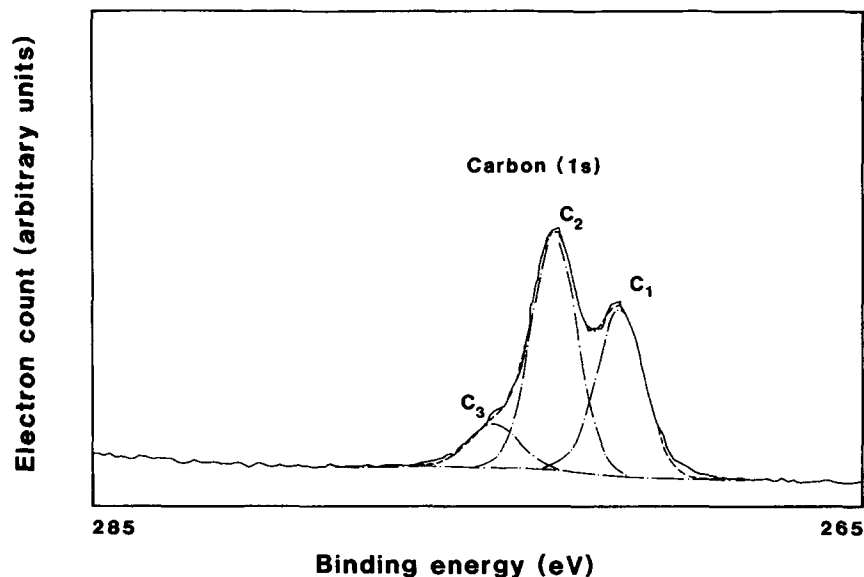


Figure 6 ESCA carbon (1s) spectrum of the topside of newsprint sample F.

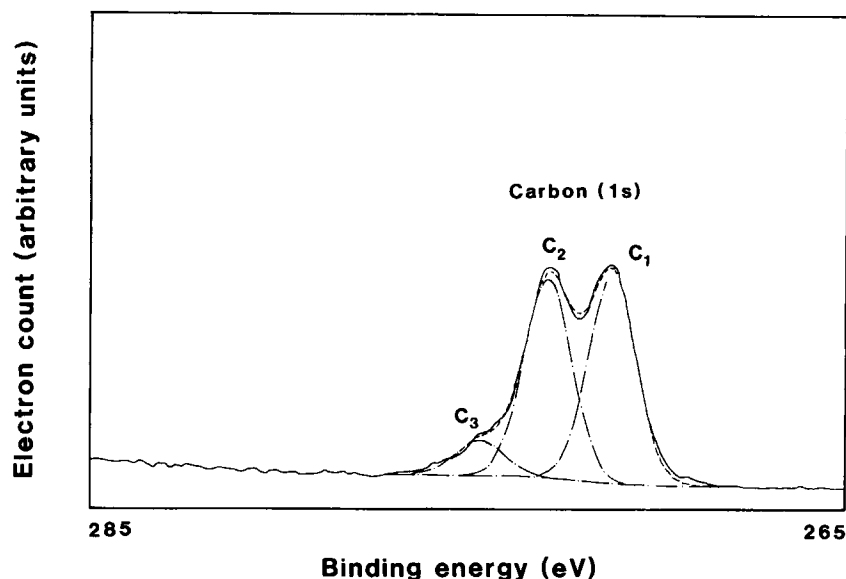


Figure 7 ESCA carbon (1s) spectrum of the topside of newsprint sample G.

these range from volatile compounds to oils to waxes and solids. Thus, it might be expected that the components may affect friction in different ways. It is even conceivable that certain compounds raise friction. For example, Broughton and Gregg show that addition of rosin size to handsheets made from sulphite pulp results in an increase in the coefficient of dynamic friction (paper against copper).¹

Unfortunately, ESCA cannot provide any insight into the proportions of the various extractives at the surface since these all have similar O/C ratios. To study the effect of different types of extractives, newsprint samples were dipped in solvent (dichloromethane) containing various concentrations of pure extractive components. The coefficient of static friction (paper against paper) was determined with an Instron tester.

Table IV Effect of Corona Discharge Treatment on the Coefficient of Static Friction of Newsprint

Treatment Time (s)	Coefficient of Static Friction ^a	Oxygen/Carbon Atom Ratio
0	0.41	0.39
5	0.53	—
30	0.58	—
300	0.59	0.47

^a Paper against paper.

The first experiment was to determine how the dipping of newsprint into dichloromethane affects friction. The results shown in Table V indicate that the procedure did not affect the friction of newsprint, suggesting that neither the surface topography was changed nor were existing surface contaminants dissolved away. The first contaminants to be tested were two fatty acids. At room temperature, oleic acid is an oil and stearic acid is a solid. Intuitively, one would expect these materials to provide some lubrication. From Table V it can be seen that both fatty acids reduce the static friction of newsprint, with oleic acid being a slightly better lubricant. Next, two higher-molecular-weight waxy materials were tested. Both are commonly found in fresh wood, but decrease in quantity on storage as they are broken down into lower-molecular-weight materials. The triglyceride glycerol trioleate behaved in a similar manner to the fatty acids, reducing the static friction by 5–18%, depending upon the concentration used. However, cholesteryl stearate had a dramatic effect, reducing friction by 36%. The last two contaminants tested were abietic acid and β -sitosterol. Abietic acid was shown to significantly increase friction at the two dichloromethane solution concentrations tested; β -sitosterol was also found to increase friction. We do not know why these compounds increase friction, but work on the lubrication of polymers suggests that the shear properties of the boundary lubricant may be important⁵; thus, it is conceivable that differences in the rheological properties of the various

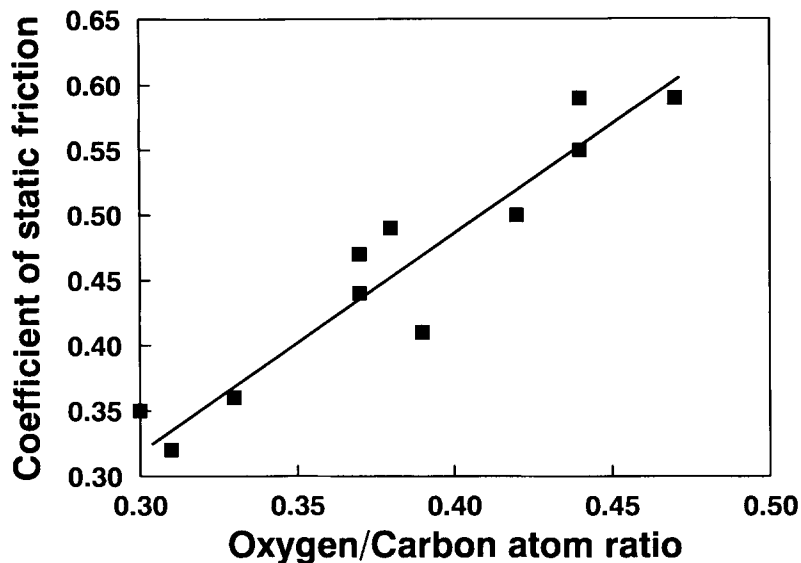


Figure 8 Plot of the coefficient of static friction (paper against paper) as a function of the oxygen/carbon atom ratio for a variety of newsprint samples from Tables III and IV.

extractive components applied may account for the different trends observed in the friction data.

These results show that individual extractive components affect paper static friction differently when applied in their pure state. However, the effect of variations in the chemical composition of surface

extractives in natural mixtures is probably more muted, as indicated by the strong correlation between static friction and surface extractives content inferred from the O/C atom ratio (see Fig. 8).

CONCLUSION

This work demonstrates that for paper samples of similar surface roughness the coefficient of friction is controlled by the degree of hydrophobicity of the sheet surface. This arises from variation in surface extractives content or the extent of surface oxidation reactions. Electron spectroscopy for chemical analysis is a valuable tool for obtaining a semiquantitative measure of the amount of extractives present on the paper surface.

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Table V Effect of Pure Surface Contaminants on the Coefficient of Static Friction of Newsprint

Contaminant	Concentration of Contaminant in Dichloromethane (mol/L)	Change in Coefficient of Static Friction ^a (%)
	Paper not dipped into solvent	
None		0
None	0.000	0
Oleic acid	0.001	-14
	0.100	-13
Stearic acid	0.001	-7
	0.010	-9
Glycerol trioleate	0.001	-5
	0.100	-18
Cholesteryl stearate	0.001	-36
β -Sitosterol	0.001	1
	0.010	24
Abietic acid	0.001	14
	0.100	74

^a Paper against paper.

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