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Load and Temperature Dependence of Frictional Force and Frictional Electrification in Friction Between Polymer Films Obtained by Simultaneous Measurements

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Synopsis

Simultaneous measurements of frictional force (F) and frictional electrification charge density (σ) were carried out for PS/Ny, PC/PET, and PS/PC polymer film sample systems in the temperature range of 10° to 100°C under various loads (W). Load dependence of F and σ were expressed by $F = \alpha W^n$ and $\sigma = \beta W^m$, respectively. Values of n and m changed with the number of rubbings and temperature. Rate of decrease in n became larger at about 90°C for all sample systems. For PS/Ny systems, the values of m and n changed in a similar manner. This behavior was explained in terms of variations in true contact area. Values of F and σ were at maxima and/or began to increase at the temperatures corresponding to the glass transition temperature or the α-peak temperature in viscoelastic polymers. The maxima of F and σ may appear when the frequency of thermal motion of molecules taking part in the adhesion and separation process coincides with the frequency of mechanical motion of adhesion and separation (friction) between two surfaces, namely, when the true contact area becomes maximum.

INTRODUCTION

Friction and frictional electrification have been studied by many workers and thought to be surface phenomena. From the point of view that the frictional electrification is an electrostatic phenomenon occurring in a friction process, it is correct to say that factors which affect the friction contribute also to the frictional electrification. Cunningham[1] found that the charge density increased with increasing the work done against the frictional force and approached a limiting value for large values of work. Nikonova et al.[2] reported that the effect of sliding velocity on the coefficient of friction was very similar to that on the frictional electrification. Sasaki[3] showed that the normal force dependence of frictional electrification was very similar to that of friction. Further, he showed that the temperature dependence of static charge was related closely to the temperature dependence of the mechanical properties of polymeric materials.[4] In order to investigate the analogous behavior of these two phenomena, the simultaneous measurement of the frictional force and the frictional electrification should be carried out on the same sample systems.

In the present paper, simultaneous measurements of the frictional force and the frictional electrification were carried out for polymer film/polymer film sample systems, and the frictional load and temperature dependence of these two phenomena were investigated.

EXPERIMENTAL

Apparatus

All the experiments were carried out using the same apparatus as stated in the literature.[5] A polymer film (polymer base) was wound around the earthed copper cylinder with a hole in the surface, over which another polymer film (polymer belt) was wrapped with a contact angle of $\pi/2$. A hole of the same size as that in the cylinder surface was made in the polymer base at the position corresponding to the hole in the cylinder. A load (W) was hung at one end of the polymer belt, and the other end was connected to a load cell. When the cylinder rotated, friction began and the electrostatic charge was generated. The generated charge on the lower surface of the polymer belt was observed as an induced voltage through the hole by an electrode set inside the cylinder. Using the calibration curve,[5] the surface charge density (σ) was easily obtained. The frictional force (F) was calculated by the equation $F = T_f - W$, where T_f is the tension in the polymer belt measured with the load cell. Friction speed (speed of circumference of the cylinder) was 1.2 mm/sec. Temperature range was 10° to 100°C and relative humidity (R.H.) was maintained at 30%.

Viscoelastic behavior (loss modulus E'') was measured by a Vibron DDV-II (Toyo-Sokki Co. Ltd.) at a frequency of 110 Hz and a heating rate of 1°C/min in the temperature range of 20° to 160°C.

In this paper, the average values of F and σ obtained in five to ten runs of experiments were shown. Within the temperature and load ranges used in the present work, macroscopic elongation of the polymer belt was not observed.

Sample Systems

Methanol-washed commercial polystyrene (PS), nylon 6 (Ny), poly(ethylene terephthalate) (PET), and polycarbonate (PC) films were used as samples. Film thickness was 0.03 to 0.05 mm. A sample system was described as polymer belt/polymer base; when PS film was used as a polymer belt and Ny as a polymer base, the system was described as PS/Ny. Each sample system was used continuously without exchanging to the extent possible for a series of experiments. However, when the surface of films was destroyed noticeably, the films were replaced.

Glass transition temperatures obtained by the DTA method at a heating rate of 10°C/min were 99°, 43°, 84°, and 148°C for PS, Ny, PET, and PC, respectively.

EXPERIMENTAL RESULTS AND DISCUSSION

Effect of Load on Frictional Force and Frictional Electrification

In order to investigate the effect of load on the frictional force and the frictional electrification, F and σ after 1, 5, 10, and 20 rubbings (one rubbing corresponds to one rotation of the cylinder) were measured in the temperature range of 20° to 100°C under the various loads. In Figures 1, 2, and 3, log F–log W and log σ–log W plots are shown for PS/Ny (30°C), PC/PET (30°C), and PS/PC (40°C), respectively. Direct lines were drawn by the method of least squares. From

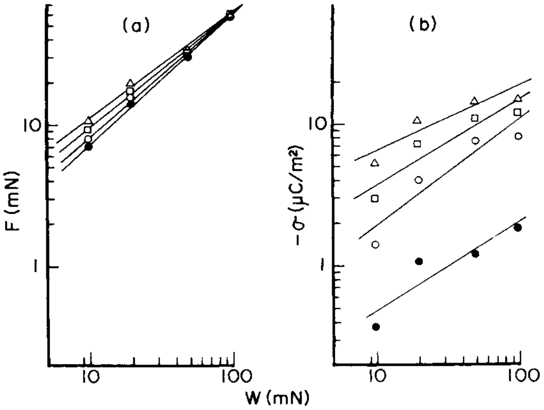

Fig. 1. Load dependence of (a) frictional force and (b) frictional electrification charge density in PS/Ny at 30°C, 30% R.H. Number of rubbings: (●) 1; (○) 5; (□) 10; (△) 20. The same notation is used in the following figures.

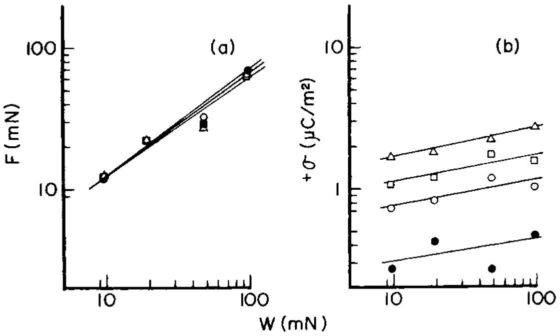

Fig. 2. Load dependence of (a) frictional force and (b) frictional electrification charge density in PC/PET at 30°C, 30% R.H.

these figures, it is clear that log F and log σ are proportional to log W in the load range used.

It was shown that the normal pressure (P) dependence of the frictional force in the sliding friction of polymers fits eq. (1):

$$\log F = \text{const.} + n \log P \qquad (\text{or } F = \alpha P^n), \qquad (1)$$

and n ordinarily takes a value between 1 and ⅔ according to the deformation mechanism of the surface asperity.[6] Further, eq. (1) was derived theoretically by considering the normal pressure dependence of true contact area (S), $S = \gamma P^n$, on the basis of the adhesion theory of friction.[6]

In the capstan method of friction as adopted in the present work, normal pressure at a given contact angle between belt and base is inconsistent with a load hung at one end of the belt. However, our results showed that there is a linear relationship between log F and log W, even if the load is used instead of the normal pressure:

$$\log F = \text{const.} + n \log W \qquad (\text{or } F = \alpha W^n). \qquad (2)$$

Equation (2) shows that in the capstan method of friction, the relationship $S = \gamma W^n$ can be also satisfied.

On the other hand, if the essential origin of the frictional electrification is the charge transfer through the surfaces in true (molecular) contact, the electrostatic charge may depend also on the contact area or on the load, namely, eq. (3) should be obeyed:

$$\log \sigma = \text{const.} + m \log W \qquad (\text{or } \sigma = \beta W^m) \tag{3}$$

In practice, from our results it was seen that log σ is proportional to log W and the true contact area plays an important role in the frictional electrification. The similar tendency between log σ and log P was found by Montgomery et al.[7] and Sasaki.[3]

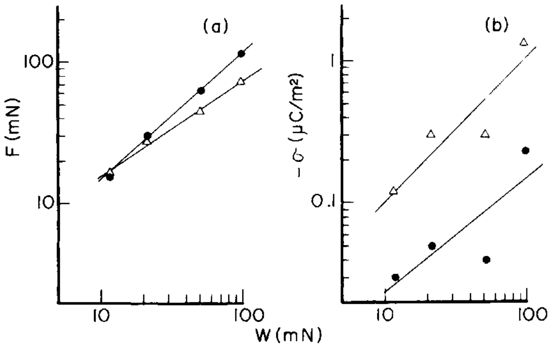

Fig. 3. Load dependence of (a) frictional force and (b) frictional electrification charge density in PS/PC at 40°C, 30% R.H.

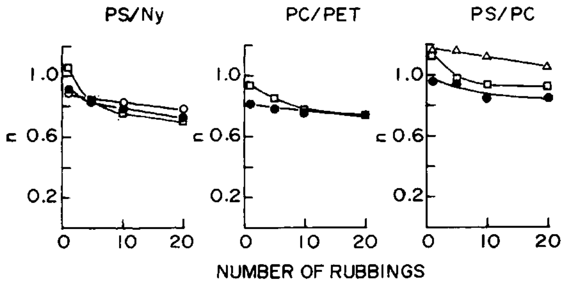

Fig. 4. Effect of number of rubbings on values of n in PS/Ny, PC/PET, and PS/PC: (●) 30°C; (○) 50°C; (△) 80°C; (□) 90°C.

Values of n and m are different for different combinations of polymer films. Numerical values of n and m calculated from the experimental data were cited in Tables I, II, and III. At a constant temperature, the value of n, which characterizes the load dependence of frictional force, decreased for all sample systems with an increase in the number of rubbings and approached a constant value (Fig. 4). This tendency appeared at each temperature. For example, the value of n in PS/Ny at 30°C changed from about 1 to ⅔ (0.92, 0.86, 0.79, and 0.72 for 1, 5, 10, and 20 rubbings, respectively). It is difficult to decide whether this tendency arose from the attainment of homogeneous contact after several rubbings or from the change of the deformation mechanism of the asperity.

If we consider the results on the basis of the adhesion theory of friction, this seems to be explainable by the change of deformation mechanism, i.e., as the

TABLE I

Numerical Values of n and m at Different Temperatures and Numbers of Rubbings in PS/Ny Sample System

No. of rub-bings	20°C		30°C		40°C		50°C		60°C		70°C		80°C		90°C		98°C	
	n	m	n	m	n	m	n	m	n	m	n	m	n	m	n	m	n	m
1	0.86	—	0.92	0.63	0.93	0.59	0.89	0.48	0.90	0.22	0.95	—	0.97	—	1.06	0.39	1.16	0.58
5	0.08	0.26	0.86	0.76	0.91	0.69	0.93	0.35	0.83	0.16	0.82	—	0.78	—	0.84	0.37	0.87	0.56
10	0.72	0.14	0.79	0.60	0.84	0.57	0.82	0.34	0.83	0.11	0.71	—	0.70	—	0.75	0.32	0.83	0.52
20	0.56	0.07	0.72	0.44	0.79	0.44	0.80	0.26	0.78	0.06	0.69	—	0.65	—	0.71	0.31	0.83	0.50

TABLE II

Numerical Values of n and m at Different Temperatures and Numbers of Rubbings in PC/PET Sample System

No. of rub-bings	20°C		30°C		40°C		50°C		60°C		70°C		80°C		90°C		100°C	
	n	m	n	m	n	m	n	m	n	m	n	m	n	m	n	m	n	m
1	0.82	0.23	0.81	0.17	0.85	0.58	0.77	0.69	0.74	0.35	0.74	0.22	0.70	0.67	0.93	0.40	1.02	0.81
5	0.79	0.34	0.78	0.20	0.76	0.52	0.73	0.80	0.72	0.58	0.70	0.45	0.72	0.79	0.85	0.57	0.86	0.70
10	0.82	0.35	0.75	0.22	0.79	0.56	0.72	0.78	0.74	0.65	0.67	0.51	0.63	0.81	0.78	0.55	0.76	0.69
20	0.82	0.28	0.75	0.24	0.75	0.52	0.72	0.80	0.72	0.71	0.53	0.48	0.61	0.73	0.73	0.51	0.65	0.60

TABLE III

Numerical Values of n and m at Different Temperatures and Numbers of Rubbings in PS/PC Sample System

No. of rub-bings	20°C		30°C		40°C		50°C		60°C		70°C		80°C		90°C	
	n	m	n	m	n	m	n	m	n	m	n	m	n	m	n	m
1	1.06	0.47	0.96	0.99	0.92	0.86	1.24	0.73	1.31	—	1.25	—	1.16	0.93	1.13	1.05
5	0.96	0.83	0.95	1.26	0.82	0.87	1.15	0.94	1.29	—	1.26	—	1.16	1.01	0.97	0.64
10	0.97	0.88	0.85	1.31	0.82	0.84	1.12	0.83	1.24	—	1.20	—	1.12	1.00	0.93	0.51
20	0.97	0.88	0.85	1.25	0.66	1.71	1.02	0.69	1.17	—	1.10	—	1.05	0.94	0.92	0.43

process of transition from the plastic deformation ($n = 1$) to the elastic one ($n = 2/3$). However, the temperature of 30°C is not high enough to produce the plastic deformation. At 70°C, the value of n for the same sample system changed also from 0.95 to 0.69. This temperature is higher than the glass transition temperature of Ny. In this case, the friction process may make the molecules in the surface layer orientate toward the shear force (plastic deformation); and after a large number of rubbings, the orientation of molecules is completed and deformation becomes elastic. This circumstance may have some influence on the extent of the contact area. As shown in Figure 4 and the tables, the initial slope of decrease of n became larger abruptly at about 90°C for all sample systems. This suggests that the variation of the deformation mechanism or extent of contact occurs more readily at higher temperatures. Further, in the temperature dependence of n in PS/Ny, n peaked at about 40°C and began to increase at about 80°C (Fig. 5). It is interesting to note that these temperatures are approximately equal to the glass transition temperatures of the materials. However, in the other sample systems, clear relationships between n and temperature could not be observed.

The behavior of m in PS/Ny was analogous to that of n: m decreased with an increase in the number of rubbings at constant temperature, and temperature dependence of m at a constant number of rubbings was quite similar to that of n (Fig. 5). However, in the other sample systems, parallel behavior between n and m could not be observed. It is interesting that the value of n was always larger than that of m for all sample systems under the same conditions.

As stated above, it was found that log F and log σ depend linearly on log W in the friction between polymer films. Load dependence of frictional force and frictional electrification may be explained by considering the increase in true contact area between polymer films. Further, the load dependence of contact area was reflected on the values of n and m under the condition of constant temperature and number of rubbings. If we consider the load dependence of

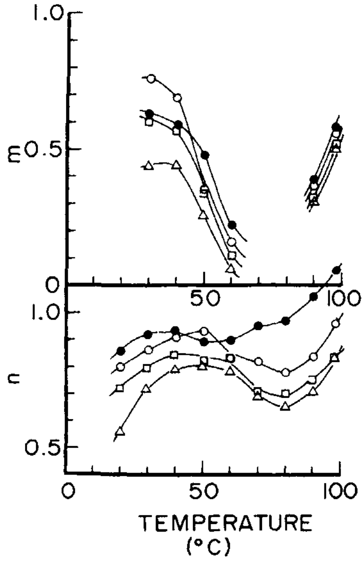

Fig. 5. Temperature dependence of n and m in PS/Ny.

electrostatic charge in this sense, we can expect that the value of m to be approximately equal to that of n at the same conditions of friction under the assumption that the frictional electrification is influenced only by the contact area. However, the value of n is always larger than that of m. Therefore, it is concluded that the static electrification is affected not only by the contact area (or load) but also by other factors, i.e., the electric conductivity of polymers, the charge distribution on the surfaces of films, etc.

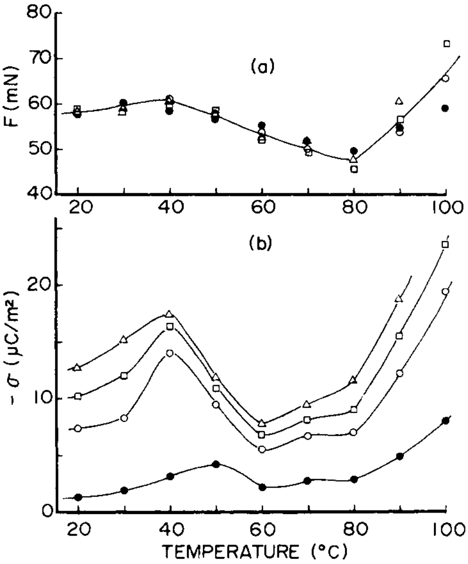

Fig. 6. Temperature dependence of (a) frictional force and (b) frictional electrification charge density in PS/Ny at W = 98 mN.

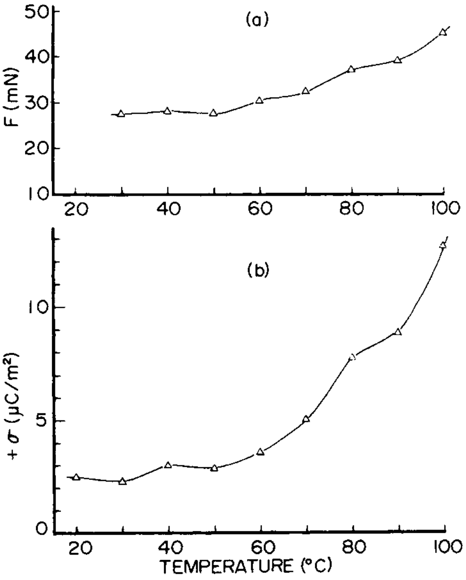

Fig. 7. Temperature dependence of (a) frictional force and (b) frictional electrification charge density in PC/PET at W = 49 mN.

Temperature Dependence of Frictional Force and Frictional Electrification

Simultaneous measurements of F and σ were carried out at various temperatures in order to investigate the effects of temperature on the frictional force and the frictional electrification. Results for PS/Ny ($W = 98$ mN), PC/PET ($W = 49$ mN), and PS/PC ($W = 49$ mN) are shown in Figures 6, 7, and 8, respectively. The number of rubbings were 1, 5, 10, and 20.

As shown in Figure 6, in the temperature dependence of the frictional force in PS/Ny, a peak was observed at about 40°C and the frictional force began to increase at about 80°C. The frictional force also began to increase at about 70°C in PC/PET and PS/PC. The frictional charge varied in the same way as the frictional force; but in the case of PS/PC, a peak was observed at about 80°C. Analogous behavior between the frictional force and the frictional electrification was observed under a comparatively high load and after a large number of rubbings. The existence of a peak and an increase in the static charge with temperature cannot be explained by the temperature variation of electric conductivity of polymers.

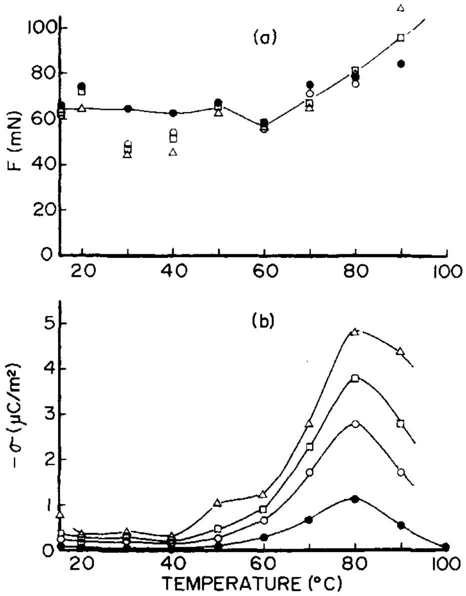

Fig. 8. Temperature dependence of (a) frictional force and (b) frictional electrification charge density in PS/PC at $W = 49$ mN.

Grosch[8] and Tabor et al.[9] studied the temperature and friction speed dependence of the coefficient of friction of polymers and showed that the viscoelastic properties of polymers affect the coefficient of friction. According to Grosch, the frictional behavior of a rubber sliding at various velocities and temperatures on a given surface can entirely be described by a single master curve and the glass transition temperature of the material, and the coefficient of friction is at maximum at the frequency corresponding to that where the loss modulus E'' of the material becomes maximum. In this case, the frictional force consists of molecular adhesion force. In the present work, temperature and friction speed ranges were not wide enough to state precisely the effect of viscoelastic properties of bulk polymers. However, it is interesting to note that in each *F*–temperature and σ–temperature curve, a peak appeared and/or an increment, which suggests the existence of another peak at a higher temperature. In order to examine this temperature, the loss modulus and the glass transition temperature of the materials were measured, and the results of the former were cited in Figure 9 (those of the latter were stated earlier). For PS, Ny, and PET, α-peaks were observed at 118°, 90°, and 114°C, respectively. In PC, no peak was observed between 40° and 110°C.

In the present case, the frictional force and the frictional electrification were observed between two polymer films; thus, in the obtained results, properties of both polymers should be reflected. As previously stated, there was a peak at about 40°C in the temperature dependence of the frictional force of PS/Ny. The increase in the frictional force was found at 80° and 70°C in PS/Ny and PS/PC, respectively. Comparing the two cases, it may be concluded that the peak at 40°C arose from the change of property of Ny because the peak at 40°C disappeared in PS/PC. This temperature seems to correlate with the glass transition temperature of Ny. It is also possible to attribute this temperature to the α-peak temperature in the loss modulus–temperature curve of Ny, although the α-peak of Ny at 110 Hz appeared at 90°C. Similarly, the rise of the frictional force at 80°C in PS/Ny and at 70°C in PS/PC is attributed to the change in the thermal or the viscoelastic properties of PS and the increase at 70°C in PC/PET to those of PET. Therefore, it is concluded that the frictional behavior of polymers is influenced by thermal or viscoelastic properties of the materials: The friction speed affects the peak position of the frictional force

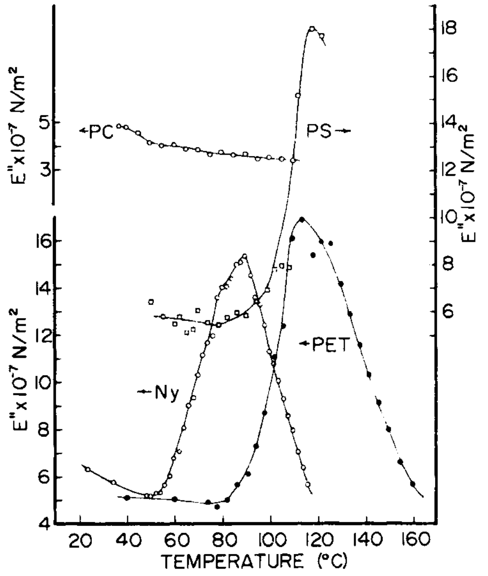

Fig. 9. Temperature dependence of loss modulus of PS, Ny, PET, and PC films.

in a similar way as the heating rate affects the glass transition temperature, or the frequency affects the peak temperature of loss modulus. Also, the peak positions shift somewhat, depending on the method of combination of the materials against a given surface.

The same discussion is possible in explaining the temperature dependence of the static electrification charge density because the behavior in static electrification is comparable with the frictional force and the thermal or viscoelastic properties of polymers (Figs. 6, 7, and 8). Hata et al.[10] reported that the static charge generated by the peeling of polymer films from many substrates was at a maximum at the glass transition temperature of polymers. Sasaki[4] found that the charge generated by friction was at a maximum at the temperature corresponding to the α-peak temperature of polymers.

As stated above, the results show that (1) there is equivalent behavior between the frictional force and the frictional electrification against the load and temperature, and (2) this behavior is affected by the changes of the thermal or the dynamic viscoelastic properties of polymers. This may be explained on the basis of the adhesion theory of friction. In this case, frictional force is caused by the adhesion force between the molecules existing throughout the true contact area. The extent of contact between polymer molecules may become maximum when the frequency of thermal motion of molecules taking part in the adhesion and separation process coincides with the frequency of mechanical motion of adhesion and separation (friction) between two surfaces.[8] Therefore, when the contact area becomes maximum, frictional force and frictional electrification become maximum. The temperatures where the maxima of the frictional force and the static charge are observed should also be influenced by the friction speed. The friction speed dependence of these phenomena is now under investigation in our laboratory.

CONCLUSIONS

Simultaneous measurements of the frictional force (F) and the frictional electrification charge density (σ) between polymer films were carried out under various conditions of load (W) and temperature, and the following results were obtained:

1. There are relationships of the type of $F = \alpha W^n$ and $\sigma = \beta W^m$ between F, σ, and W.
2. Generally, n is larger than m. The value of n decreases when the number of rubbings are increased. The rate of decrease in n becomes larger at higher temperatures. The value of m behaves in a similar manner as n for a certain combination of polymers.
3. Values of F and σ are at maximum or begin to increase at the glass transition temperature or at the α-peak temperature. Similar behavior between F and σ is explained in terms of variation of the true contact area. The frictional electrification is affected not only by the electric property of the polymer, but also by thermal or mechanical factors.

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