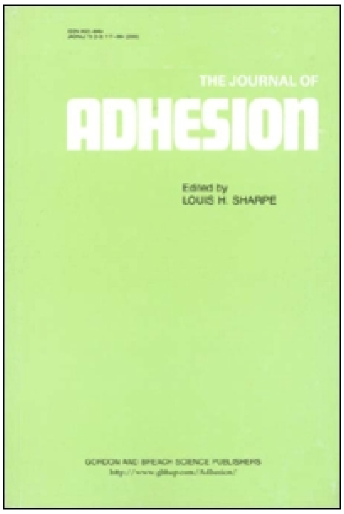

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Tack and Viscoelasticity of Block Copolymer Based Adhesives[†]

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Tackification of block copolymers of the linear ABA or radial $(AB)_x$ type, in which the center block (B) is in the rubbery state at use temperature, follows principles established for other rubbery polymers except for peculiarities of the systems introduced by the block polymer domain structure. Successful pressure-sensitive adhesives result wherever the tackifer is compatible with the rubbery phase, which forms the continuum. Connectivity of the hard phase generally leads to loss in compliance, making tack ultimately contact-limited; whereas tack is low, adhesion can be developed by control of dwell time, temperature and contact pressure. A previously explored criterion for contact limitation, defining a critical minimum 1-second creep compliance, appears to be valid for block polymer-based adhesives, as is the rule that an effective tackifier must raise T_g (in the present case of the rubbery continuum) while also acting as a plasticizer.

Although viscoelastic processes play an exceedingly important role in the tack phenomenon, quantitative correlations for block polymer adhesives are difficult to establish because of the thermorheological complexity and multiphase morphology of these systems. Nevertheless, some semiquantitative relations have been developed. They are, in addition to the contact criterion, a correlation of tack and dynamic loss modulus and a criterion for tackifier compatibility with the rubbery phase.

INTRODUCTION

Anyone who has handled a piece of pressure-sensitive adhesive tape has an intuitive grasp of the phenomenon of "tack". Webster's dictionary defines it as "the quality or state of sticking or adhering; adhesiveness; stickiness". A little additional thought reveals it as a complex property, involving a bonding and a debonding process carried out in fairly rapid succession. In the bonding stage the objective is that maximum molecular contact be established between the soft adhesive and the microscopically rough substrate. Subsequent debonding is essentially an adhesion test, the outcome of

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which will depend on the extent of contact established in the bonding stage, the strength of the intermolecular forces acting at the adhesive/substrate interface and the rheological properties of the adhesive layer. (In many cases the substrate may be considered to be infinitely rigid.)

A criterion for the establishment of contact on a microscopic scale was proposed several years ago by Dahlquist.[1] It states that the compressive creep compliance on the time scale of the bonding process (*ca.* 1 sec) should be of the order of 10^{-7} cm^2/dyne or larger, *i.e.*

$$D(1) \geqq 10^{-7} \text{ cm}^2/\text{dyne}. \tag{1}$$

When $D(1)$ is substantially smaller, serious loss in tack will result from limitation of contact. Clearly, we have here the first crucial influence of a viscoelastic property of the adhesive on the tack phenomenon.

In the debonding step we are dealing with the adhesion of a soft viscoelastic material to a rigid surface. This basic problem has been studied extensively in the literature, most recently by Gent, Andrews and Kinloch.[2-4] These authors employed the concept of a characteristic adhesive failure energy, Θ, defined as the change in free energy of deformation with the length (area) of a growing crack at the interface, to demonstrate the role of viscoelastic processes in the mechanics of adhesive failure. Specifically, Andrews and Kinloch[3] were able to show that

$$\Theta = \Theta_0 F(\dot{c}a_T) \tag{2}$$

where Θ_0 is an "intrinsic failure energy" which, in absence of covalent bonding, is closely related to the thermodynamic work of adhesion. The function F represents the viscoelastic contribution by the adhesive, $\dot{c}$ being the crack propagation velocity and a_T the time–temperature shift function of linear viscoelasticity.[5] Although tack is generally not measured in test piece geometries conducive to evaluation of Θ and the resulting tack-values will not necessarily be *simply* related to Θ, Eq. (2) can be used to gain useful insight into tack and tackification. Since F is a monotonically increasing function of $\dot{c}a_T$, a quantity which in turn increases strongly in moving from the rubbery region of viscoelastic response toward the transition region, it is possible to attain reasonable values of Θ even when Θ_0 is small, *i.e.* with non-polar adhesives relying entirely on dispersion forces for bonding to the substrate. Equation (2), of course, also makes clear that the chemical nature of the substrate and its interaction with the adhesive will have an effect on adhesion and tack.

High molecular weight rubbers, block polymers included, have little or no tack because first of all the contact criterion (Eq.1) is not satisfied. It can, of course, be met by the addition of a plasticizer or low molecular weight diluent. Plasticizers function in two ways. They loosen up the entanglement network[6] and usually decrease segmental friction. However, in the present situation one would like to *increase* segmental friction and hence the value of F. This can be done by using a soluble low molecular weight resin of substantially higher T_g than that of rubber, *i.e.*, a tackifier. The tackifier does loosen up the entanglement network and so increases the compliance in the entanglement plateau and terminal regions, but it also raises T_g and so moves the use temperature of the adhesive composition closer to its transition region. These relationships are illustrated schematically in Figure 1. Note how at the use temperature the polymer does not satisfy Eq. (1), but the tackified composition does.

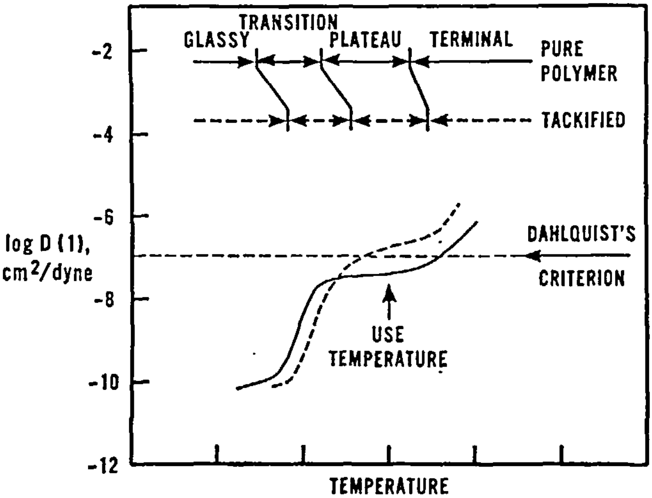

FIGURE 1 One second creep compliance vs. temperature of a high molecular weight amorphous polymer before (——) and after (- - - -) incorporation of a tackifier.

The above analysis is also valid for block polymer-based adhesives, although some additional factors become important. The block polymers of principal interest here are the linear and radial tri- and multiblock polymers of butadiene or isoprene and styrene: SBS or SIS and $(SB)_x$ or $(SI)_x$, where $x > 2$. These polymers are well known to undergo microphase separation forming domain structures. It has been shown that their outstanding advantage over homopolymers or random copolymers is holding power, derived from the persistence of the domain structure in the tackified adhesive compositions.[7] The polystyrene domains act as multifunctional physical cross-links and filler particles and so greatly diminish shear creep at long times. The tackifier thus should be incompatible with the polystyrene domains, but compatible with the rubbery polydiene phase. Moreover, the latter should be the *only* continuous phase, since connectivity of the polystyrene domains greatly decreases the compliance in the temperature–time region of interest, resulting in failure to meet the contact criterion. Fortunately, the use of large quantities of polydiene-compatible tackifiers tends to prevent polystyrene domain connectivity, such as results from cylindrical or lamellar morphologies. Still, formulation of adhesives of highest tack becomes difficult with block polymers containing more than 30% styrene.[7]

The present work is a continuation of earlier studies from this laboratory on tackification of diene-styrene block polymers.[7,8] It examines some of the relationships developed in more detail and addresses itself to some previously uninvestigated questions.

EXPERIMENTAL

Materials

The block polymers used were commercial radial-structure block copolymers manufactured by Phillips Petroleum Company (Table I).

TABLE I

Polymer characteristics[a]

Polymer	Composition	$M_w/1000$	$M_n/1000$
Solprene†418	85 :15 isoprene/styrene	300	220
Solprene†417	80 :20 butadiene/styrene	190	150
Solprene†416	70 :30 butadiene/styrene	140	110
Solprene†414	60 :40 butadiene/styrene	130	100

[a] Nominal properties.

† A trademark of Phillips Petroleum Company.

Tackifying resins were likewise of commercial manufacture. Their chemical type and origin are listed in Table II.

TABLE II

Resin	Type	Source
Foral 85	Rosin ester	Hercules, Inc.
Wingtack 95	Polyterpene	Goodyear Chemical Co.
Super Sta-Tac 80	Polyolefin	Reichhold Chemical Co.
Zonarez 7085	Polydipentene	Arizona Chemical Co.
Zonarez B-85	Poly(β-pinene)	Arizona Chemical Co.
Picco Alpha 115	Poly(α-pinene)	Hercules, Inc.
Kristalex 1120	Poly(α-methylstyrene)	Hercules, Inc.

Adhesive formulations and testing

A simple basic formulation was used throughout this study:

Polymer	100
Tackifier	as specified
Stabilizer†	1

Variable tackifier concentration series were prepared with Solprene 418 using Foral 85 and Wingtack 95, respectively. In addition the various resins were evaluated at 100 phr concentration in both Solprene 418 and 417. Solprene 416 and 414 were used with 100 phr Foral 85 only.

Formulations were mixed 30 minutes at 50 rpm in a Brabender Plasticorder under a nitrogen blanket. The mixing chamber was heated with oil circulated from a bath maintained at 180°C.

The pressure-sensitive films for the tack measurements were prepared by a hot melt molding procedure. Small portions of the mix were pressed between Teflon-coated foil and 2-mil Mylar backing with platens at 150°C to form films. Shim stock was used to control the film thickness at 0.06 ± 0.01 mm. The probe tack values of these films were determined one day after preparation.

Tack measurements were made exclusively with a Polyken Probe Tack Tester, Testing Machines, Inc., Amityville, N.Y.: test results reported are averages taken from five specimens. Contact time was 1 second, contact pressure 100 grams/cm^2, and separation rate 1 cm/second. Unless otherwise stated the test temperature was 25 ± 1°C with a polished stainless steel probe.

Probe tack determinations at temperatures above 25°C were obtained by enclosing the probe, specimen, and specimen carrier in a heated chamber. Specimens were placed in the heated chamber 20–30 minutes before testing.

Viscoelastic measurements

Dynamic viscoelastic measurements were carried out in shear, using a Rheovibron Model DDV-II Viscoelastometer (Toyo Instruments Co., Japan). The test specimen was a 3 mm square of 0.4–0.5 mm thickness sandwiched between brass plates. Creep measurements were carried out at 22°C in tension on $0.16 \times 0.685 \times 4$ cm strips. A constant applied stress of 3.3×10^5 dynes/cm^2 was used in all creep determinations. The creep of the adhesive formulations was not strictly linear. However, non-linearity was modest; typically, a 2.5-fold increase in stress caused a 20% increase in the measured compliance. A correction was applied for diminishing cross-section during the creep experiment. Details of this calculation are to be published elsewhere.

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† Irganox 1010.

RESULTS AND DISCUSSION

Creep compliance

Testing the Dahlquist[1] contact criterion requires knowledge of the 1-second creep compliance (Eq. 1). Unfortunately, $D(1)$ is not easily measured directly, the time being too short after the (hypothetically) instantaneous application of stress. Creep compliances can be calculated from dynamic measurements, but these are usually performed at higher frequencies and so furnish values of $D(t)$ at shorter times only. In the present study both types of measurements were combined to obtain $D(1)$ by interpolation. For times longer than 10 seconds creep was measured directly (in tension) and values of $D(t)$ at times shorter than 0.1 sec. were calculated from dynamic shear data by the approximation method of Riande and Markovitz.[9]

$$J(t) \cong \{[J'(\omega)]^2+[J''(\omega)]^2\}^{\frac{1}{2}}, \; t = \omega^{-1} \tag{3}$$

and the well known relation

$$3D(t) = J(t) \tag{4}$$

applicable to incompressible materials—a good assumption for soft rubbery adhesives. Here ω is the angular frequency in radians/sec, J' and J'' are the dynamic storage and loss compliances in shear and $J(t)$ the shear creep compliance.

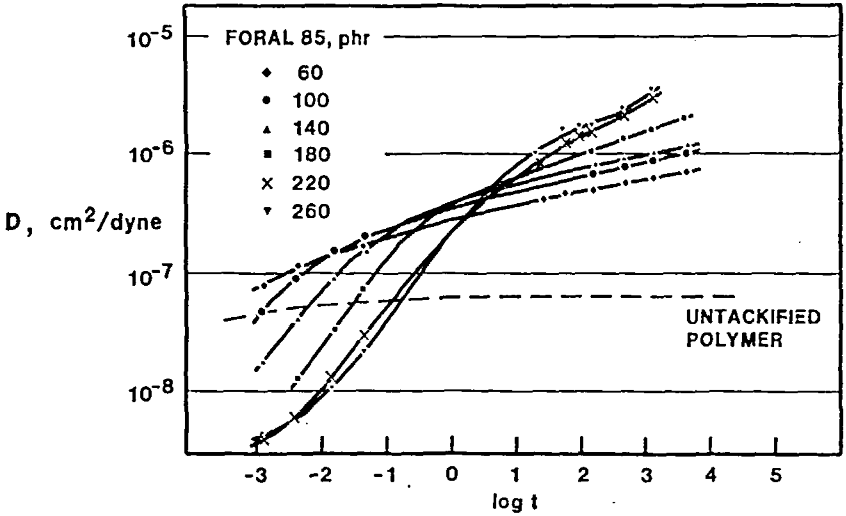

FIGURE 2 Creep compliance at 22°C for isoprene-styrene block polymer (Solprene 418) at varying level of rosin ester tackifier.

Figure 2 shows $D(t)$ at 22°C for a series of adhesives formulated from Solprene 418 and Foral 85 tackifier, the level of the latter increasing from 60 to 260 phr in 40 phr increments. The response of the untackified block polymer is seen to be extremely flat. This reflects the highly elastic character of the block polymer resulting from the low T_g of its polyisoprene matrix and effective cross-linking by its polystyrene domains. Addition of tackifier increases the creep compliance at long times, the effect of a low molecular weight diluent. At very short times, however, $D(t)$ is decreased, because the tackifier raises T_g. In isothermal data this results in a shift of the transition zone toward longer times so that the more highly tackified compositions are now well inside the transition region from glassy to rubbery behavior.

At intermediate times there are multiple cross-overs. With the exception of the pure polymer, which has little tack, $D(1)$ lies comfortably above 10^{-7} cm²/dyne. Table III shows that all six adhesive compositions have satisfactory tack. Also listed are the locations of the maximum in loss modulus at a fixed frequency of 35 Hz, illustrating the shift of the glass transition temperature of the rubbery phase, and the dynamic moduli at room temperature.

TABLE III

Effect of tackifier level on probe tack of Solprene 418 based adhesives

Tackifier	Level (phr)	$T(G''_{\max})$ (°C)	$G' \times 10^{-7}$ (dynes/cm²)	$G'' \times 10^{-7}$ (dynes/cm²)	$D(1) \times 10^{7}$ (cm²/dyne)	Probe tack (grams)
Foral 85	60	−22	0.27	0.095	2.7	1060
	100	−12	0.28	0.24	3.5	1010
	140	−4	0.48	0.94	3.9	1300
	180	14	1.80	2.55	3.8	1560
	220	24	2.55	2.05	1.8	1560
	260	33	4.60	1.75	1.8	1490
Wingtack 95	60	−26	0.20	0.064	3.1	740
	100	−3	0.28	0.23	4.4	1170
	140	12	0.95	1.09	3.5	1440
	180	24	2.30	2.40	2.2	650

Similar data for Solprene 418/Wingtack reveal the same general trends, except that tack exhibits a sharper maximum with resin concentration. Creep curves for these compositions are shown in Figure 3.

Figure 4 shows the creep behavior of Solprene 418 with five effective tackifiers at constant concentration (100 phr). The curves show remarkable similarity and all compositions again satisfy the Dahlquist criterion. Tack values are shown in Table IV, together with data for poly (α-methylstyrene) resin which is not a tackifier and which clearly does not meet the $D(1) > 10^{-7}$ criterion.

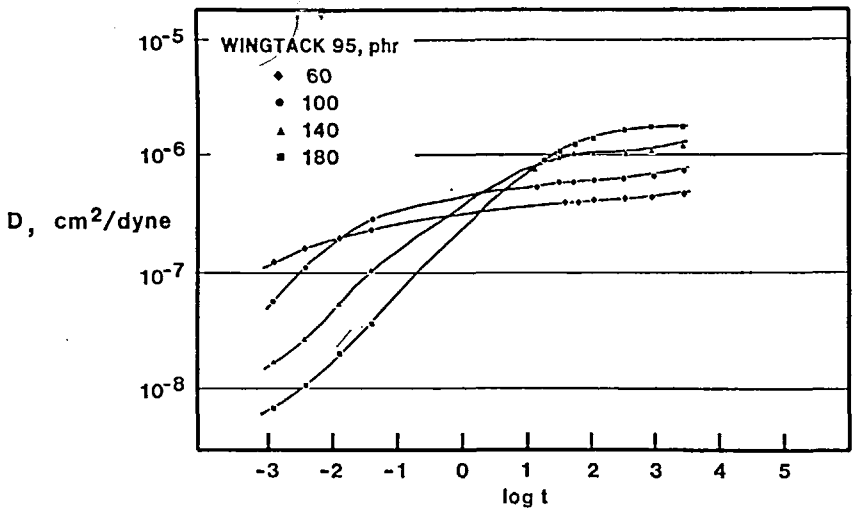

FIGURE 3 Creep compliance at 22°C for isoprene-styrene block polymer (Solprene 418) at varying level of polyterpene tackifier.

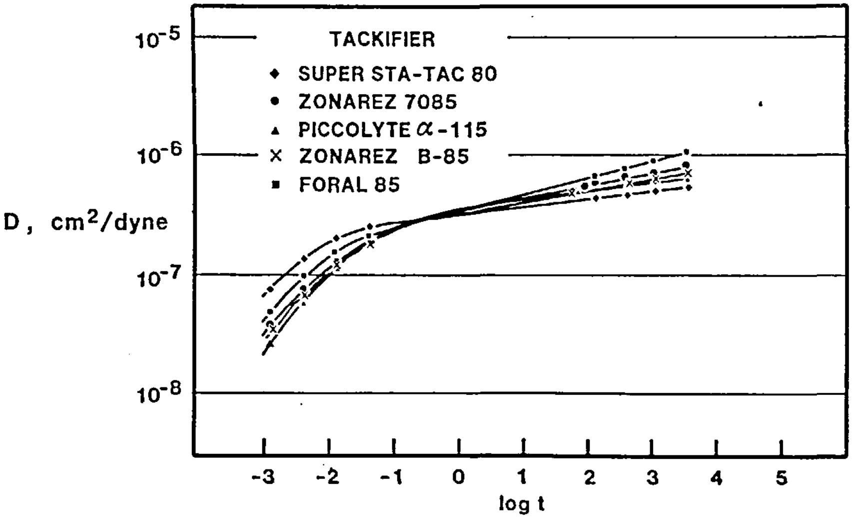

FIGURE 4 Creep compliance at 22°C for isoprene-styrene block polymer (Solprene 418) tackified with 100 parts of various resins.

TABLE IV

Comparison of tackifiers in Solprene 418 (100 phr)

Tackifier	$T(G''_{max})$ (°C)	$G' \times 10^{-7}$ (dynes/cm^2)	$G'' \times 10^{-7}$ (dynes/cm^2)	$D(1) \times 10^7$ (cm^2/dyne)	Probe tack (grams)
Wingtack 95	−3	0.28	0.23	4.4	1170
Super Sta-Tac 80	−14	0.23	0.135	3.0	1090
Foral 85	−12	0.28	0.24	3.5	1010
Zonarez 7085	−2	0.34	0.34	3.4	1520
Picco Alpha 115	−10	0.38	0.46	3.6	1540
Zonarez B-85	−2	0.38	0.35	3.3	1120
Kristalex 1120	—[a]	8.80	1.70	<0.1	0

[a] Less than −40°.

Contact criteria†

In Ref. 7 a contact criterion, more conservative than the one of Dahlquist, was formulated in terms of the storage modulus at 35 Hz:

$$G'(35\text{ Hz}) < 4 \times 10^6 \text{ dynes/cm}^2 \qquad (5)$$

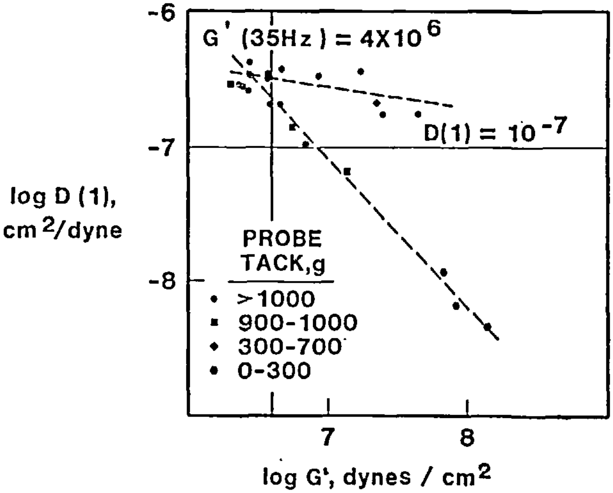

FIGURE 5 Comparison of contact criteria.

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† Contact criteria are, of course, dependent on contact pressure, dwell time and surface roughness. The criteria discussed here are for the experimental conditions of the present study.

Figure 5 shows a mapping of $D(1)$ and G' (35 Hz). The data are taken from Tables III and IV and similar tests on butadiene-styrene block polymers (see Experimental). In the upper left quadrant both criteria are met and the probe tack value exceeds 700 g in all cases (all squares and circles). The Dahlquist criterion is seen to be remarkably good, with only minor violations near the critical compliance of 10^{-7} cm^2/dyne.

The data of Figure 5 appear to group themselves along two lines. The points clustering about the dashed line are all for 100 phr of tackifier, with polymer and tackifier varying, while those along the dotted line represent variable tackifier levels. These are *not* fundamental correlations. They merely reflect the fact that for the two variable tackifier concentration series the compliance curves lie close together at $t = 1$, while the short-time compliances and the dynamic moduli (at $\omega = 1/t$) vary appreciably with concentration (Figures 2 and 3). For the adhesives at constant tackifier level the shapes of the creep curves are more nearly similar, hence the compliances vary inversely to the moduli.

Criterion for tackifier effectiveness in Diene-Styrene block polymers

Kraus and Rollmann[8] have shown that the value of the dynamic storage modulus of block polymer-based adhesives of the present type at the minimum in tan δ, located between the polydiene and polystyrene domain glass transitions (see inset of Figure 6), is approximately given by

$$G'(\tan \delta_{\min}) = v_2^2(\rho/M_e)RT(1+2.5\,c+14.1\,c^2) \tag{6}$$

if the tackifier is soluble in the rubbery continuum, but not in the polystyrene domains which are assumed spherical. Here v_2 is the volume fraction of *polymer* in the polydiene phase, c the volume fraction of polystyrene domains in the entire composition, M_e the polydiene entanglement spacing in the undiluted polymer, ρ the density, R the gas constant and T absolute temperature. If a prospective tackifying resin is compatible with and "alloys" itself with the polystyrene domains v_2 will remain unity, c will be increased by the tackifier and, very likely, some connectivity of the glassy phase will develop if the level of tackifier is high enough. Hence G' will greatly exceed the value calculated by Eq. (6). The same will be true if the resin is compatible with neither block and forms a third phase. In neither case will tack of the composition be enhanced. In this manner Eq. (6) becomes a criterion of suitability of a resin as a tackifier, to which must be added the condition that the resin increase T_g.

Among the adhesives used to demonstrate the validity of Eq. (6) were some of the compositions of the present paper. It was observed that the equation holds almost quantitatively for pure block polymers ($v_2 = 1$), but that the v_2^2 concentration dependence appears to overestimate the modulus.

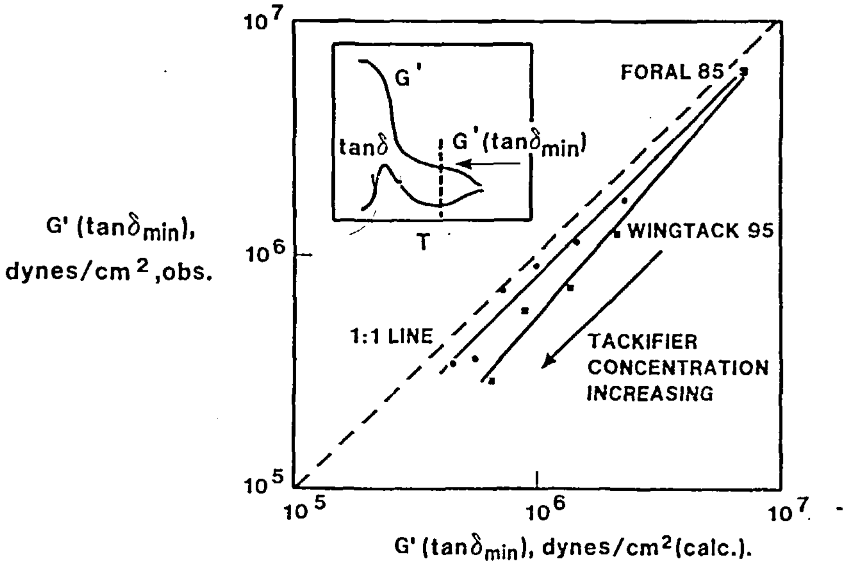

FIGURE 6 Observed vs. calculated storage moduli (35 Hz) at temperature of minimum in loss tangent. Data for Solprene 418.

The two tackifier concentration series (Table III) permit closer examination of the problem. As seen from Figure 6, the difference between observed and calculated $G'(\tan\,\delta_{\min})$ indeed increases with concentration, suggesting a stronger dependence than v_2^2. However, it is obvious that the two tackifiers do not follow exactly the same concentration law. The best-fit exponents are 2.28 and 2.62, respectively, and their use would force very good agreement between calculation and experiment. However, this does not appear to be a particularly useful exercise. Instead, we note that Eq. (6) is independent of the nature of the diluent; it only considers loosening of the entanglement network and the reinforcing effect of the polystyrene domains. Moderate specific effects would, of course, be entirely conceivable. On the other hand, the substantial increases in T_g of the rubbery phase, caused by large amounts of tackifiers, shift the minimum in tan δ into the region approaching the polystyrene domain glass transition. Some softening of polystyrene domains is, therefore, possible; Eq. (6) treats them as perfectly rigid. The fact that Wingtack 95 raises the position of the minimum in tan δ more than Foral 85 would be consistent with this interpretation (Figure 7). Interestingly, the position of the tan *δ-maximum*, accompanying the glass transition of the rubbery phase, is the same for the two tackifiers when the comparison is made at equal volume fraction. In spite of all this the degree of success of Eq. (6) in predicting $G'(\tan\,\delta_{\min})$ for a series of highly complex materials without the use of any adjustable parameters is quite satisfactory.

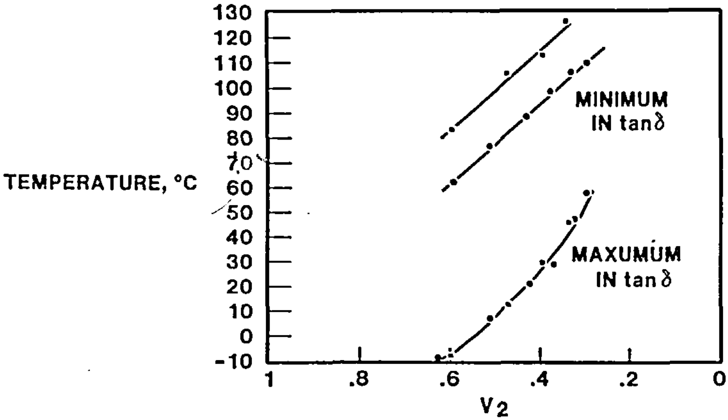

FIGURE 7 Positions of maxima and minima in loss tangent (35 Hz). Solprene 418 with Foral 85 (●) and Wingtack 95 (■).

The probe tack value

To explain different values in the probe tack test with various block polymer-based adhesives Kraus *et al.*[7] attempted a correlation with the loss modulus at 35 Hz. It has been estimated[1,10] that the time scale of the debonding step in the probe tack test is of the order of 0.01 sec which is roughly comparable to a dynamic test at 35 Hz, as $t = 1/\omega = 1/2\pi(35) = 0.005$ sec.† Moreover, the loss modulus was chosen since it is proportional to energy dissipation and energy losses around the tip of growing cracks are well known to figure prominently in both cohesive and adhesive failure phenomena.[2,3]

The result of this attempt was a curve with a maximum in tack at a loss modulus near 10^6 dynes/cm^2. The declining portion of the curve was ascribed to increasing degrees of contact limitation, as the storage modulus increases concurrently to values well above the contact criterion of Eq. (5). However, as shown above this criterion is rather conservative and contact limitation was not actually proven in the earlier study. To provide such proof use may be made of the fact that the tack of a contact-limited adhesive should respond to increasing dwell time and contact temperature. Table V shows an example.

Of the three adhesives, C fails both contact criteria (Eqs. 1 and 5), A passes and B appears marginal. Only C responds significantly to increasing dwell time and temperature. For the others the tack value decreases with temperature, as expected. Indeed at 100°C all three adhesives gave tack values of

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† Frequencies available on the Rheovibron apparatus are 3.5, 11, 35 and 110 Hz. An exact match to 0.01 sec. could have been obtained by interpolation, but this would not have affected the results significantly.

100–200 g irrespective of contact time. To further illustrate these effects experiments were made in which the temperatures of the bonding and debonding steps were varied independently (Table VI). It is clear that adhesive C, while exhibiting very little tack at 25°C, gives excellent adhesion if the bonding temperature is sufficiently high.

TABLE V

Effects of dwell time and temperature on tack

	A Solprene 417 -Foral 85[a]	B Solprene 416 -Foral 85[a]	C Solprene 414 -Foral 85[a]
$D(1)\times 10^7$, cm^2/dyne, 25°C	2.2	0.7	0.12
$G'\times 10^{-7}$ dynes/cm^2, 25°C, 35 Hz	0.45	1.38	6.9
$G''\times 10^{-7}$ dynes/cm^2, 25°C, 35 Hz	0.18	0.79	1.8
Probe Tack, g			
25°, 1 sec. dwell	960	1120	70
5 sec. dwell	980	1250	170
50°, 1 sec. dwell	790	640	430
5 sec. dwell	840	690	760

[a] 100 phr.

TABLE VI

Effect of bonding and debonding temperatures[a]

Bonding Temp. (°C)	Bonding Time (sec.)	Debonding Temp. (°C)	Probe Adhesion or Tack (grams)
25	1	25	70
50	1	50	430
50	1[b]	25	700
100	1	100	190
100	1[b]	25	1440

[a] Adhesive C of Table V, debonding rate 1 cm/sec.
[b] Plus time to cool to 25°C.

Figure 8 combines the points of the ascending branch of Figure 17 of reference 7 with the data of the present study on all adhesives satisfying the Dahlquist contact criterion. The result is a satisfactory, if somewhat scattered, correlation. The only serious violation occurs for the Solprene 418/Wingtack 95 adhesive containing 180 phr of tackifier (see Table IV). The correlation is, of course, limited on the side of large values of G''. Because these are rubbery adhesives, whose tan δ will generally not exceed unity, G'' cannot increase indefinitely without also increasing G', leading to eventual contact limitation of tack. In any case, the maximum probe tack possible with these adhesives appears to be in the vicinity of 1600 g.

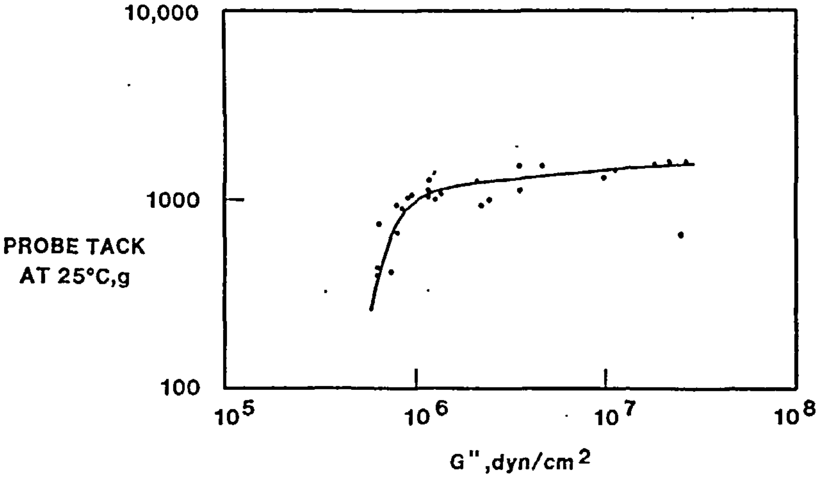

FIGURE 8 Tack vs. 35 Hz loss modulus.

The correlation of Figure 8 does not consider the possibility of an effect on tack of the magnitude of the intermolecular forces acting at the interface. Since the chemical composition of the adhesives varies appreciably (the data cover butadiene/styrene and isoprene/styrene block polymers of different styrene contents with different tackifying resins at several concentrations) some of the scatter could easily be due to this variable. In fact, the success of the correlation suggests that these interactions are roughly constant. Since the adhesives are essentially non-polar substances, the interaction with the (stainless steel) substrate is likely to be entirely by dispersion forces, which do not vary strongly among different hydrocarbon materials. That there is a significant contribution to tack from surface interactions is easily demonstrated by changing the probe material.

Tack vs. probe material

Table VII shows tack values for Solprene 418/Foral 85 (100 phr) adhesive against six different probe materials. The tack of this adhesive under the present conditions is not contact-limited. With only minor, probably in-significant, inversions at 50°C, tack decreases in the same order for the different materials at all temperatures. The fact that the organic polymers, but also aluminium, show increasing tack with rising temperature is not clearly understood. For the polymers one might visualize some (very limited) mixing at the interface, but this could hardly apply to aluminium.

TABLE VII

Tacks as function of substrate material

Probe material	Probe Tack, g 25°C	 35°C	 50°C
Stainless steel	1280	1080	820
Glass	1010	980	860
Aluminium	410	590	600
Polyphenylene sulfide	280	360	630
Polyamide	170	150	270
Polytetrafluoroethylene	40	30	90

CONCLUSIONS

The results of this and earlier studies[7,8] illustrate clearly the close connection between tack and viscoelasticity for block-polymer based pressure-sensitive adhesives. The correlations found seem valid, but they are essentially semi-quantitative and limited to certain aspects of the problem. A general theory of tack and adhesion seems a long way off, particularly for such complicated materials as block polymers. Foreseeable impediments are thermorheological complexity (multiphase block polymers do not follow simple time–temperature superposition), complications arising from non-linearity, and the detailed effects of the morphology on mechanical behavior. Added to this is, of course, the difficulty of the stress analysis problems involving complicated test geometries.

Of the three relationships between tack and viscoelastic properties discussed here, the Dahlquist contact criterion seems generally applicable to all pressure-sensitive adhesives. The correlation between tack and loss modulus has been demonstrated only for block polymers; it may or may not be of broader validity. The tackifier selection criterion has been tested with isolated compositions based on polyisoprene and polybutadiene homopolymers ($c = 0$ in Eq. 6) and found to apply.[8] The utility of all these correlations lies, of course, mainly in the insight they provide into the mechanism of tack. In practice, it is always easier to measure the tack of a composition directly.

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