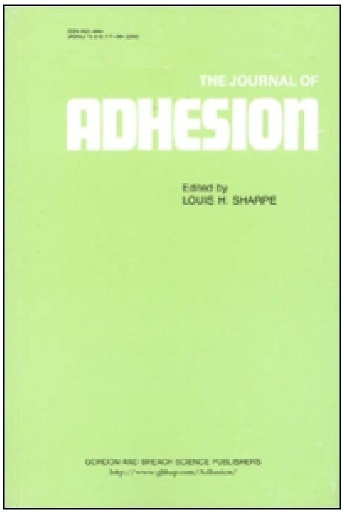
This article was downloaded by:
On: *22 January 2011*
Access details: *Access Details: Free Access*
Publisher *Taylor & Francis*
Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



The Journal of Adhesion

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

Shear Strength of Pressure Sensitive Adhesives and its Correlation to Mechanical Properties

A. Zosel[a]
[a] Kunststofflaboratorium, Ludwigshafen, Germany

To cite this Article Zosel, A.(1994) 'Shear Strength of Pressure Sensitive Adhesives and its Correlation to Mechanical Properties', The Journal of Adhesion, 44: 1, 1 — 16

To link to this Article: DOI: 10.1080/00218469408026613

URL: http://dx.doi.org/10.1080/00218469408026613

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.informaworld.com/terms-and-conditions-of-access.pdf

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

J. Adhesion, 1994, Vol. 44, pp. 1–16
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

Shear Strength of Pressure Sensitive Adhesives and its Correlation to Mechanical Properties

A. ZOSEL

Kunststofflaboratorium, BASF AG, 6700 Ludwigshafen, Germany

(Received March 1, 1993; in final form August 9, 1993)

Correlations between shear resistance and the mechanical properties of pressure sensitive adhesives are studied by measuring the deformation behaviour in the static and the dynamic shear test and determining the dynamic shear modulus of the adhesive. For polymers with low or moderate viscosities, the shear strain *vs.* time characteristics in a static shear test and, accordingly, the static shear strength, can be evaluated from the master curves of the dynamic shear modulus or the dynamic viscosity. The dynamic shear strength also can be calculated. These exact calculations cannot be applied to highly viscous or slightly crosslinked polymers. On the basis of the model experiments, empirical correlations between shear strength and the dynamic shear modulus are established which seem to be generally valid.

KEY WORDS pressure sensitive adhesives; shear strength; dynamic shear modulus; shear viscosity; viscoelasticity; polymers; model experiments; holding time.

1. INTRODUCTION

The performance of a pressure sensitive adhesive is governed mainly by three properties: tack, peel strength, and shear strength. The last property indicates the resistance of an adhesive to shearing forces and will be the subject of this study. The standard method of determining shear strength is the "static" shear test[1-3] which measures the time required to pull a defined area of a tape from a test panel under a constant load. This time is called the holding time or the time to failure, t_c, the subscript c being related to "cohesion." As the static shear test is rather time consuming, a "dynamic" test has been proposed which deforms a sandwich type (lap shear) sample, with a similar geometry to that in a static test, with a constant shear rate and determines the maximum force as a measure of shear resistance.[4]

The behaviour of an adhesive film in a shear test is governed by viscous flow and elastic, *i.e.* recoverable, deformation. Accordingly, shear strength should be, at least qualitatively, related to the viscoelastic properties of the adhesive, which, on the other hand, are influenced by the molecular structure of the polymer from which the adhesive is made. When the separation occurs by cohesive failure, the holding time can be regarded as a measure of the cohesive strength of the material.

In the case of adhesive failure only a lower limit for the cohesive strength of the adhesive can be given. This study is restricted to cohesive failure.

This paper, parts of which were presented at the PSTC "1^{st} World Congress for Pressure Sensitive Adhesive Tape," 1992,[5] mainly has two objectives. The first question to be addressed is, if it is possible to calculate the static shear strength from the viscoelastic data of the adhesive, especially from the dynamic shear modulus and the shear viscosity, and as this approach is limited to a small group of materials, as will be shown, an empirical correlation between shear strength and the shear modulus is established. The second point is a similar correlation between dynamic shear and the viscoelastic properties.

2. EXPERIMENTAL PROCEDURE, MATERIALS

2.1. Static Shear Measurements

For the objectives of this study the standard static shear tests have the disadvantage that only the "end point" of the shear *versus* time curve, the holding time t_c, is determined. We have, therefore, developed an instrument which simulates this test and measures the shear strain in the sample as a function of time during the whole experiment. A schematic diagram of this apparatus is shown in Figure 1. A constant shear force, F, applied to the sample with a test area of 10 mm × 8 mm by a dead load in the range between 20 and 500 g, and the resulting "elongation," Δl, of the sample is determined as a function of time with a linear differential transducer. The shear strain, γ, in the adhesive layer is given by $\gamma = \Delta l/d$ (d being the thickness of the adhesive layer), provided that the tensile deformation of the adhesive with its backing between the end of the steel plate and the upper clamp can be neglected. This has been achieved by reinforcing the backing by a second steel plate with a thickness of 1 mm. This steel plate also prevents the sample from bending under the influence of the applied force. The sample is placed in a heating chamber permitting measurements between room temperature and about 200°C. A similar device has been described by Miyagi and Yamamoto.[6]

The samples were prepared by applying the polymer solution or dispersion to a 30 μm thick polyester film. After drying the polymer forms a homogeneous film which has a thickness of 25 μm for most measurements in this study.

The beginning of the static shear experiment is similar to a creep measurement with a constant shear stress.[7] The test area, A, of the sample, however, decreases with rising Δl from $A_o = l_o \cdot b$ at the start, to zero at failure according to:

$$A = A_o \cdot \left(1 - \frac{\Delta l}{l_o}\right), \tag{1}$$

as is shown in the upper part of Figure 2. The decrease of A results in an increase of the shear stress, $\sigma = F/A$, in the adhesive layer, as the shearing force, F, is constant:

$$\sigma = \frac{\sigma_o}{1 - \Delta l/l_o}, \text{ where } \sigma_o = \frac{F}{A_o}. \tag{2}$$

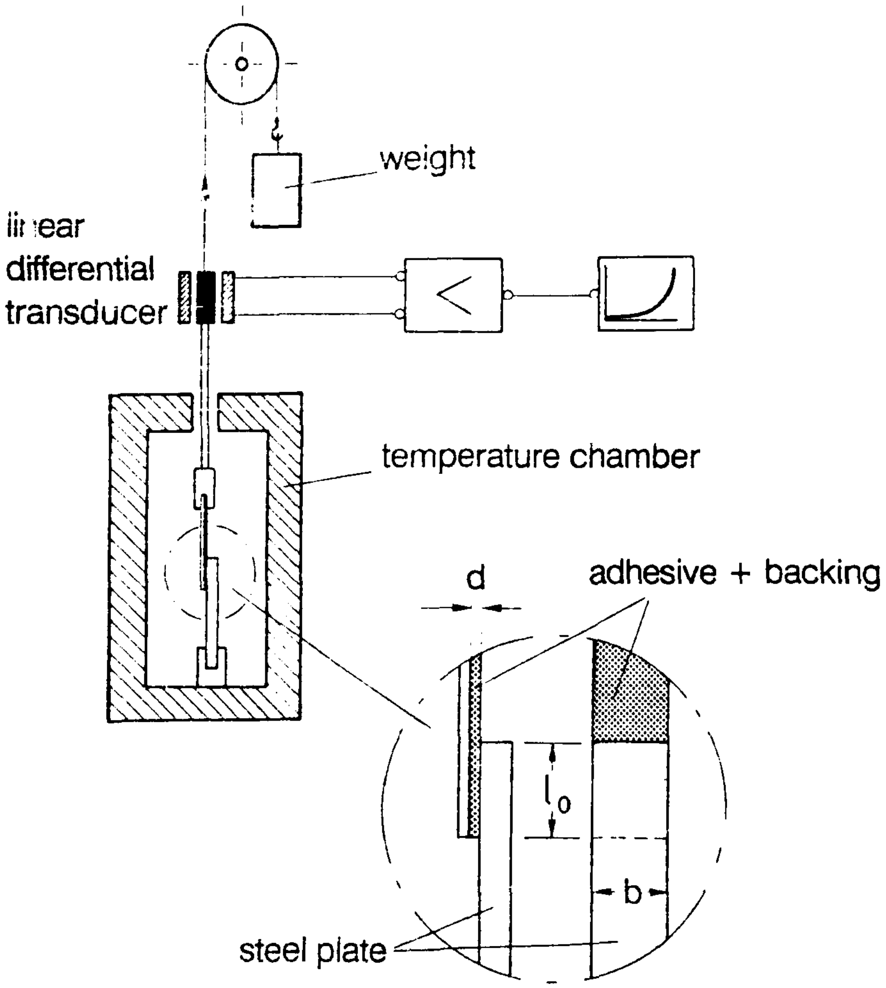


FIGURE 1 Schematic diagram of the instrument for the measurement of the shear *versus* time curve and the static shear strength.

A typical plot of Δl *versus* time, t, subsequently called the shear characteristics of the material, is shown in Figure 2. At $t=0$ the load is applied to the sample; the time when Δl is equal to l_0 corresponds to the holding time in a standard shear test. If the load is removed before failure of the sample, the non-recoverable and the elastic, *i.e.* recoverable, parts of the total strain can be determined as it is done in normal creep experiments.

The curve in Figure 2 represents the measurements of one single specimen. Measurements of a large number of similar specimens lead to a standard deviation of about 20% which seems to be mainly caused by the sample preparation. A fairly large scatter of the holding times is also known for the standard static shear tests.

In addition to the measurements with the instrument shown in Figure 1, a part of

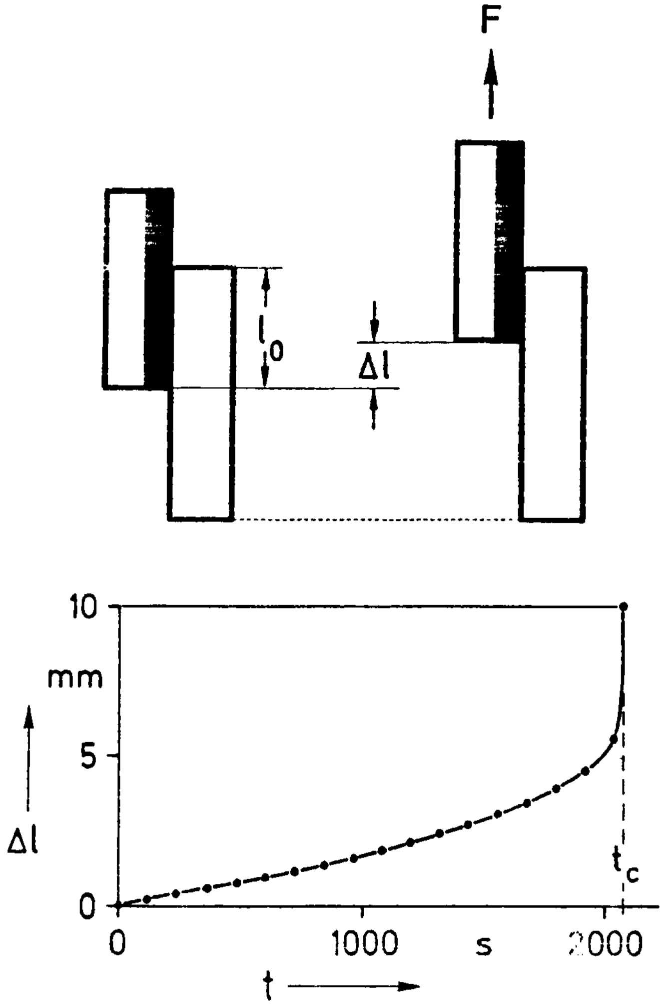


FIGURE 2 "Deformation," Δl, of the sample in a static shear test and example for a plot of the deformation *versus* time, measured with a polyisobutylene with an average molecular weight of $1 \cdot 10^5$ g/mole, shear force $F = 1.6$ N, $T = 23°C$.

the shear strength data given in this paper were obtained by standard shear tests with a test area of 20 × 25 mm and a dead load of 1 kg. The preparation of the test pieces also followed a standard procedure.

2.2. Dynamic Shear Measurements

The dynamic shear test is performed with a tensile tester (Instron tensile tester 1121). A sample similar to that shown in Figure 1 is deformed and separated with a constant cross-head speed, v, leading to a constant shear rate

$$\dot{\gamma}_d = \frac{v}{d} \tag{3}$$

in the adhesive, where d again is the thickness of the adhesive layer. The resulting force, F, is measured as a function of time. Cross-head speeds between 0.05 and 10 mm/min, mostly 5 mm/min, were applied. As already mentioned, the maximum force, F_o, or the corresponding shear stress, $\sigma_o = F_o/A_o$, is used in order to characterize the dynamic shear strength.

The static and dynamic shear tests were performed at 23°C, with exception of some measurements shown in Figure 4.

2.3. Dynamic Mechanical Analysis (DMA)

The mechanical behaviour of the polymers is investigated with a dynamic mechanical analyzer (Rheometrics RDA II). The sample is subjected to torsion between horizontal, parallel plates, and the complex dynamic shear modulus $G^* = G' + i \cdot G''$ is determined as a function of temperature and frequency. G^* consists of two components, the storage modulus G' and the loss modulus G''. DMA is increasingly used to investigate the viscoelastic properties of pressure sensitive adhesives (e.g. Ref. 8). In this study, both moduli have been determined as a function of the angular frequency, ω, at various temperatures above T_g. The modulus *versus* frequency curves at different temperatures are superimposed to a master curve at a certain reference temperature, T_o, by use of the well-known method of frequency/temperature superposition.[7] The master curves for G' and G'' represent the viscoelastic behaviour of the polymer on a frequency scale much more extended than could be covered by measurement at a single temperature.

Figure 3a gives an example for the master curves of an uncrosslinked polymer of rather high molecular weight (polyisobutylene with $M_w = 3.8 \cdot 10^5$ g/mole, $T_o = 23°C$). Three frequency ranges with different viscoelastic behaviour can be distinguished:

—the glass transition range at angular frequencies above about 10^3 s^{-1},
—the plateau range between 10^{-3} and 10^3 s^{-1} where the moduli are determined by entanglements, and
—the terminal or flow range at low frequencies where G' and G'' decrease with decreasing frequency and viscous flow becomes dominant.

From G' and G'' the dynamic viscosity, $|\eta^*|$,

$$|\eta^*| = \frac{(G'^2 + G''^2)^{1/2}}{\omega} \tag{4}$$

can be calculated. This is shown in Figure 3b. For viscous, *i.e.* uncrosslinked polymers, the dynamic viscosity equals the steady-state viscosity, $\eta(\dot{\gamma})$, as a function of shear rate, $\dot{\gamma}$:

$$\eta(\dot{\gamma}) = |\eta^*(\omega)|, \quad \dot{\gamma} = \omega\ . \tag{5}$$

This relation has been empirically found by Cox and Merz.[9]

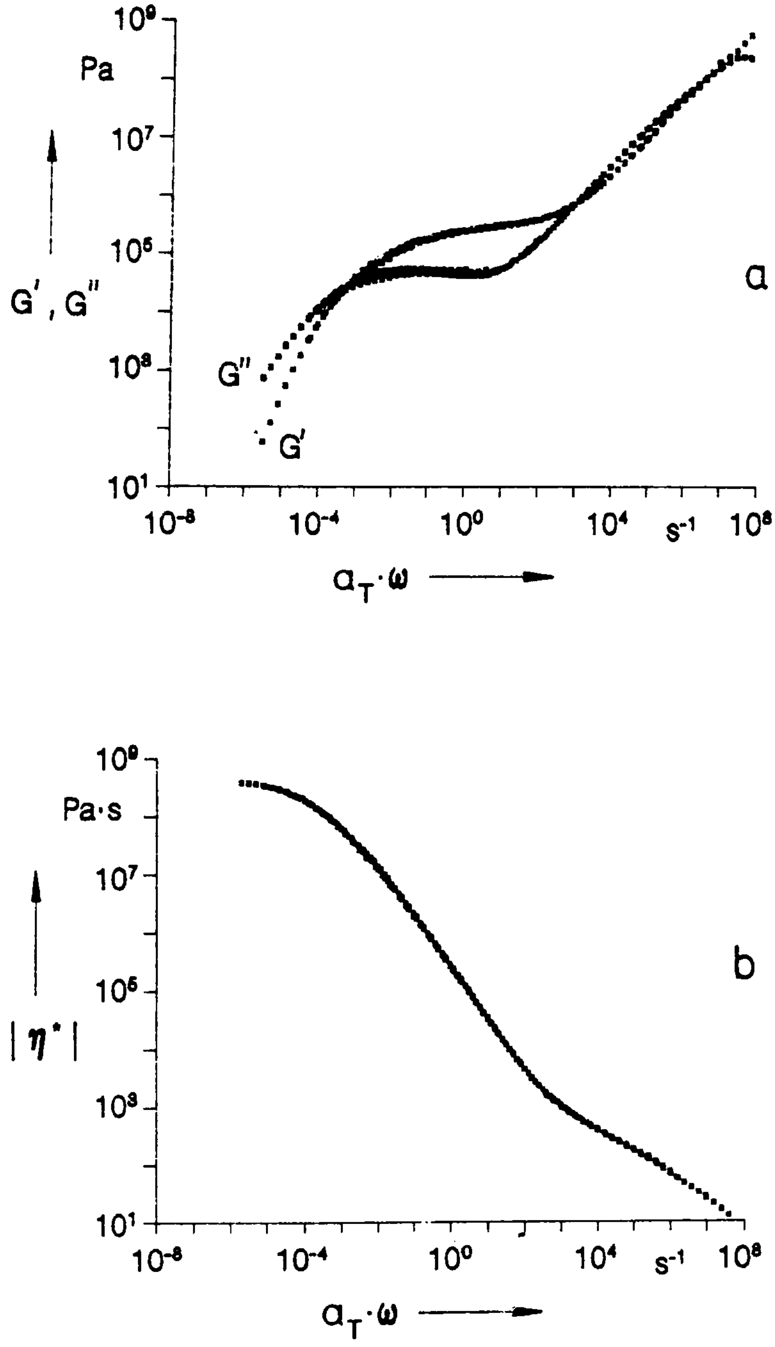


FIGURE 3 Master curves of the storage modulus, G', and the loss modulus, G''; (a) of an uncrosslinked polymer (polyisobutylene with $M_w = 3.8 \cdot 10^5$ g/mole) and dynamic viscosity $|\eta^*|$, (b) *versus* angular frequency, ω, calculated with the Cox-Merz relation (equation (4)), T = 23°C.

2.4. Materials

Various series of polymers were used as model pressure sensitive adhesives:

- —two polyisobutylenes (PIB) with weight average molecular weights of $1.0\cdot 10^5$ and $3.8\cdot 10^5$ g/mole, respectively.
- —a series of emulsion polymers of n-butylacrylate (PBA) which have been polymerized in the presence of different concentrations of a chain transfer agent (t-dodecylmercaptane, DMCT) or a bifunctional monomer (butandiol diacrylate, BDA2), in order to obtain a series of samples covering the whole range from uncrosslinked, viscous, to crosslinked, rubber-like, materials.
- —copolymers of n-butylacrylate and methacrylic acid (MAA) with MAA concentrations between 1 and 10%.

In addition, blends of polyacrylates and polyacrylate/resin blends to be used as pressure sensitive adhesives were studied.

3. STATIC SHEAR

3.1. Calculation of Static Shear Strength from the Shear Modulus for Viscous Polymers

The approach to calculate the static shear strength from the dynamic shear moduli G′ and G″ and the shear viscosity is limited to cohesive failure, as already mentioned, and to viscous, *i.e.* uncrosslinked polymers. Starting with the equation

$$\sigma = \eta \cdot \dot{\gamma} \tag{6}$$

and taking into account equation (2) for σ, which can be written as

$$\sigma = \frac{\sigma_o}{1-\gamma/\gamma_o} \text{ with } \gamma = \Delta l/d \text{ and } \gamma_o = l_o/d, \tag{7}$$

one obtains

$$\sigma_o = \eta\cdot(1-\gamma/\gamma_o)\cdot d\gamma/dt, \tag{8}$$

which, by integration, leads to the following equation for the holding time, t_c:

$$t_c = \int_0^{t_c} dt = \frac{A_o}{F} \cdot \int_0^{\gamma_o} \eta\cdot(1-\gamma/\gamma_o)\, d\gamma\,. \tag{9}$$

This equation can be solved for a constant viscosity, $\eta = \eta_o$, leading to the formula of Dahlquist[10]

$$t_c = \frac{l_o^2\, b\, \eta_o}{2\, d\, F}\,. \tag{10}$$

The calculation of t_c is also possible for a power law dependence of the viscosity, *i.e.* $\eta \sim \gamma^{-n}$.[11]

In this study we try to calculate the Δl *versus* t curve and t_c for a general shear rate dependence of the viscosity. As the integral in equation (9) cannot be solved generally, we use the following approximation:

$$t_c = \frac{A_0}{F} \cdot \sum_{\gamma=1}^{N} \eta_\nu \cdot (1 - \gamma_\nu/\gamma_0) \cdot \Delta\gamma \tag{11}$$

The following procedure is applied for the calculation of the holding time and the shear characteristics $\Delta l = \Delta l(t)$. The steady-state viscosity, η, can be calculated as a function of $\dot{\gamma}$ or σ *via* the Cox-Merz relation (equation (4)). The total shear deformation, γ_0, is divided into N intervals, $\Delta\gamma$, and for every interval an average shear stress, σ_ν, and the corresponding viscosity, η_ν, can be deduced from the η *versus* σ curve. Starting with σ_0 and $\eta(\sigma_0)$, Δl can also be calculated stepwise as a function of time, and this calculated Δl *vs.* t curve can be compared with the experimental shear characteristics. The number N of intervals has been chosen sufficiently high (typically between 10 and 40), so that the calculated Δl *versus* t curves become independent of N within a deviation of less than 3%.

In Figure 4, the calculated and the measured shear characteristics are compared for the polyisobutylene with a molecular weight of about $1 \cdot 10^5$ g mole^{-1} at two temperatures. The experimental curves fall into the hatched area between the broken lines, whilst the calculated curve is drawn as a full line. The calculated and the measured shear curves agree well at the higher temperature of 35°C, where the PIB has a zero shear viscosity $\eta_0 = 2.5 \cdot 10^5$ Pa·s. At 23°C with a higher viscosity of $\eta_0 = 8 \cdot 10^5$ Pa·s, the calculated curve falls outside the "experimental range" and the

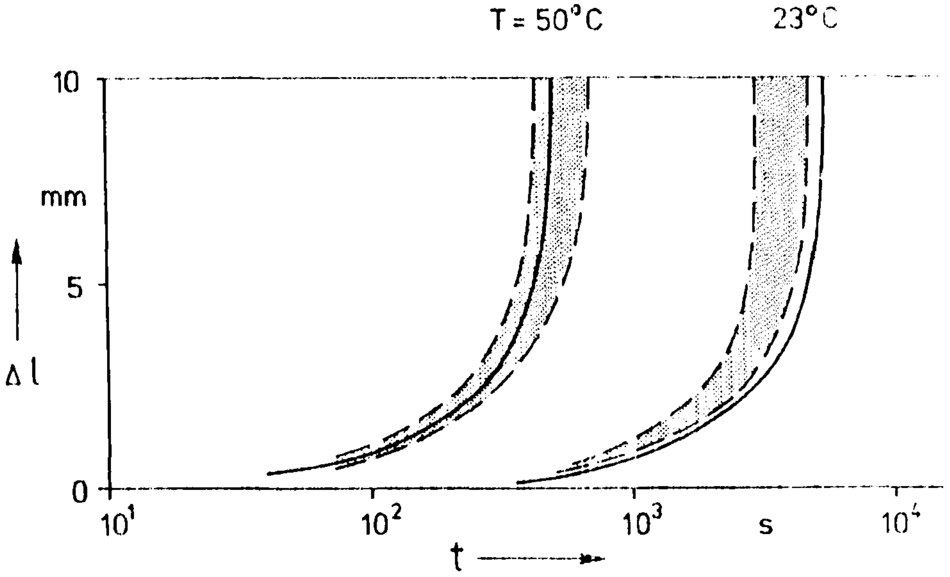


FIGURE 4 Comparison of the measured and the calculated shear *versus* time curves for polyisobutylene ($M_w = 1 \cdot 10^5$ g/mole) at 23 and 35°C. F = 1.2 N. —: calculated curve, the experimental curves fall into the hatched area.

calculated holding time, t_c, is about 40% higher than the mean value of the measured holding time.

This discrepancy becomes worse with increasing viscosity. Figure 5 shows the calculated and the measured shear characteristics for the polyisobutylene with the higher molecular mass, determined at 23°C with shear forces of 1.6 and 5 N, respectively. As the number of experimental curves is fairly small due to the very long holding times, up to 150 h, only one experimental curve is shown. The difference between calculated and measured shear characteristics is much larger for this polymer which has a zero shear viscosity of $2.5 \cdot 10^8$ Pa·s at 23°C. The difference between the calculated and the measured t_c values increases with rising shearing force and reaches nearly one order of magnitude for 5 N. Measurements of the polyacrylates, the results of which are not presented here, confirm that the agreement between calculated and measured curves is good for materials with low and moderate viscosities, whilst larger deviations are observed for higher viscosities.

Polymers near the gel point, which are highly branched or even have some gel content, and especially slightly-crosslinked samples, exhibit a different shear behaviour which is exemplified in Figure 6, showing the shear characteristics for a slightly-crosslinked polybutylacrylate, measured with 4 different loads. Two time ranges are found: creep with a small, only slightly time dependent shear strain at short times and a very strong, nearly catastrophic increase of Δl at longer times. A Δl *versus* t curve of this type cannot be calculated with the approximation, deduced above, apart from the fact that the approach leading to equation (10) assumes that viscous flow is governing the shear behaviour. The strong increase of the shear strain at longer times, shown in Figure 6, possibly indicates a new deformation and fail-

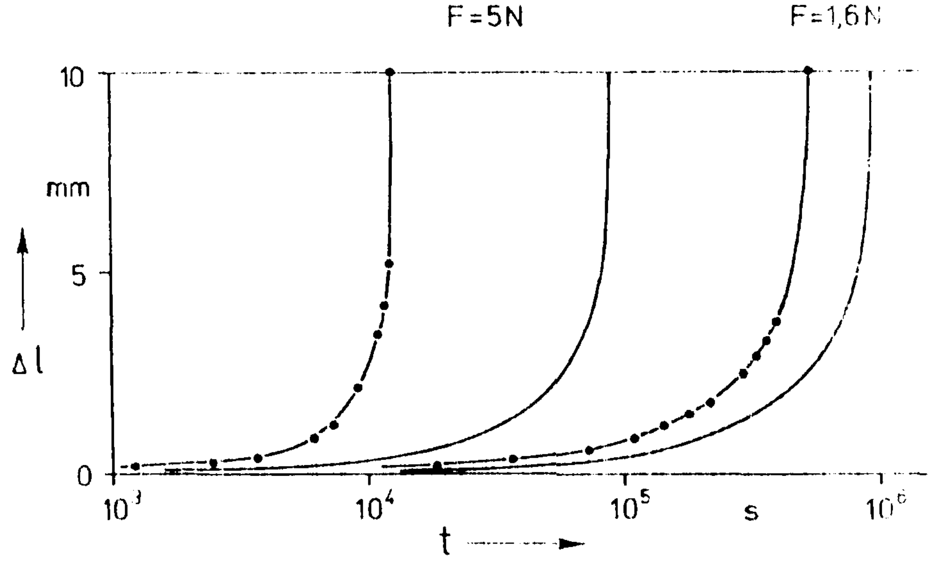


FIGURE 5 Comparison of the measured (—●—) and the calculated (———) shear *versus* time curves for polyisobutylene ($M_w = 3.8 \cdot 10^5$ g/mole). T = 23°C. F = 1.6 and 5 N.

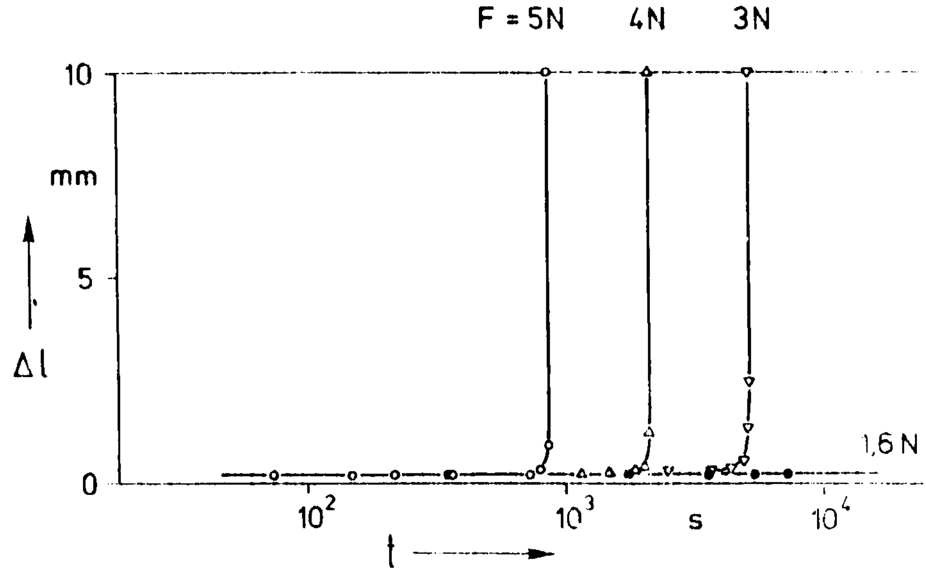


FIGURE 6 Measured shear *versus* time curves for a slightly-crosslinked polybutylacrylate, determined with 4 different shear forces, F.

ure mechanism to be operative which cannot be expected to be described by linear viscoelasticity.

It follows that the approach to calculate Δl(t) presented in the equations (6) to (11), which is based on linear viscoelasticity, can be applied neither to samples with high viscosities, *i.e.* above about $1 \cdot 10^6$ Pa·s, nor to crosslinked polymers. Highly viscous and crosslinked polymers, however, are the materials mainly used in the pressure sensitive adhesive industry. The objective of the next section, therefore, will be to establish an empirical correlation between the static shear resistance and the viscoelastic behaviour which is valid more generally and can also be applied to pressure sensitive adhesives.

3.2. Empirical Correlations between Static Shear Strength and Shear Modulus

Empirical relations of static shear strength to viscoelastic data of pressure sensitive adhesives have been reported by various authors. Krenceski and Johnson[12] correlate the holding time to the dynamic viscosity at a frequency of 0.1 s^{-1} for a series of polyisobutylenes with different molecular weights. The correlation becomes worse for samples with broad molecular weight distributions. Dale *et al.*[13] discuss relations between shear strength at room temperature and the storage modulus at 127°C, as well as the tensile strength at room temperature for acrylic pressure sensitive adhesives, finding correlation coefficients in the range of 0.74 to 0.87. Andrews *et al.*[14] deal with a correlation of shear strength to peel strength from soft-machine peel testing. The choice of the viscoelastic data, *i.e.* the frequencies and the temperatures, at which the moduli were determined, seems to be a little arbitrary and not logically established in some of these publications.

In order to establish an empirical relation between the static shear strength and the dynamic shear modulus one has to take into account that the time scale of a static shear test is characterized by the holding time, t_c. Shear tests with different holding times, accordingly, take place at different time scales and deformation rates. That means that it does not seem reasonable to correlate the static shear strength to a viscoelastic quantity such as a modulus or a viscosity at a fixed frequency. A static shear test with a long holding time is expected to be related to the modulus or the viscosity at a low frequency, whilst a shear test with a shorter time is governed by the corresponding viscoelastic quantities at a higher frequency.

At first, we define an angular frequency, ω_s, which is correlated to the static shear experiment. For this purpose, we introduce an average shear rate by the equation

$$\dot{\gamma}_s = \frac{l_o}{d \cdot t_c} \tag{12}$$

and define a corresponding angular frequency, $\omega_s = \dot{\gamma}_s$. According to Figures 4 to 6, which show that the shear rate during a static test is not constant but increases with time, it is obvious that this is only a very crude approximation which, however, may be sufficient for an empirical correlation.

In order to establish such a correlation between the holding time and the dynamic modulus, ω_s is calculated from t_c, and $|G^*(\omega_s)|$ is evaluated from the corresponding G' and G'' *versus* ω plots. $|G^*|$ is chosen for this comparison instead of G' or G'', as it is related to the dynamic viscosity, $|\eta^*| = |G^*|/\omega$, which governs the static shear test on viscous samples, as has been shown in the foregoing section.

In Figure 7, $|G^*(\omega_s)|$ is plotted against the holding time, t_c, for a number of the

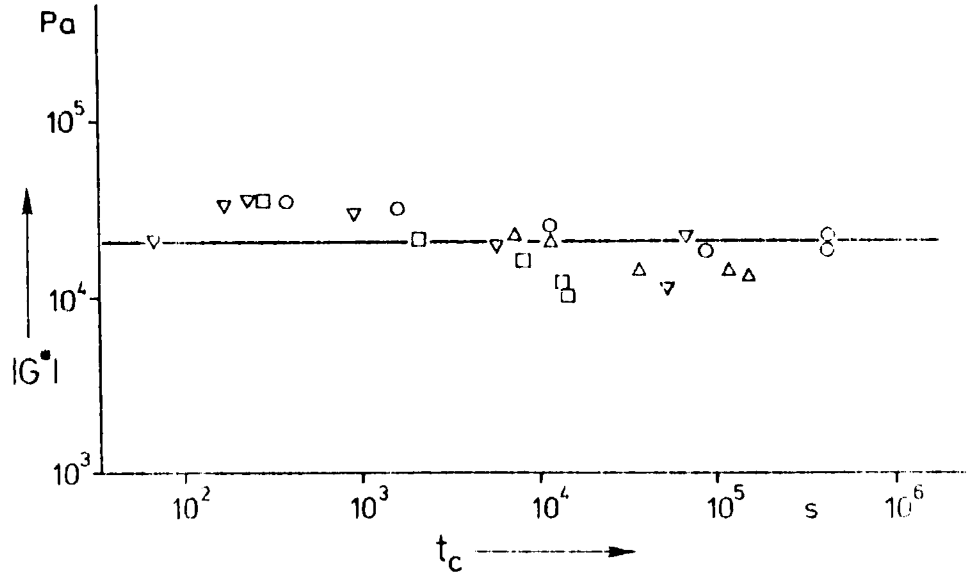


FIGURE 7 Correlation between the shear modulus, $|G^*|$, at the angular frequency, ω_s, defined by equation (12), and the measured holding time, t_c. Samples: Polybutylacrylate with DMCT (▽), BDA2 (□) and MAA (○), acrylic PSA's (△).

materials studied in this work. In this case, the holding times were obtained by a standard shear test with a test area of 25×25 mm^2 and a shear force of 10 N. It follows from Figure 7 that all moduli fall into a range between about 1 and $3.5 \cdot 10^4$ Pa with an average value of $2 \cdot 10^4$ Pa and a standard deviation of 50%, whilst t_c varies over about 4 orders of magnitude. That means that the polymers studied here have a more or less constant modulus at the frequency related to the holding time and are, accordingly, in a similar state with respect to their dynamic mechanical behaviour at frequencies correlated to the time scale of the static shear test. The relation shown in Figure 7 is also valid for other shear forces; however, with different levels of the constant shear modulus.

4. DYNAMIC SHEAR

The dynamic shear test is less complicated than the static one, insofar as the adhesive is sheared with a constant shear rate, $\dot{\gamma}_d = v/d$ (equation (3)), as already explained. Accordingly, this results in a constant shear stress

$$\sigma_o = \eta \cdot \dot{\gamma}_d \tag{13}$$

during the whole experiment for an uncrosslinked, viscous polymer for which the viscosity can again be evaluated with the Cox-Merz equation. The test area, A, decreases from the starting value, $A_o = l_o \cdot b$, to zero at the end of the test:

$$A = l \cdot b = (l_o - v \cdot t) \cdot b = A_o \cdot (1 - \Delta l / l_o), \quad \Delta l = v \cdot t \tag{14}$$

The shearing force, $F = \sigma_o \cdot A$, for a viscous material is, thus, expected to decrease linearly with increasing Δl:

$$F = F_o \cdot (1 - \Delta l / l_o), \quad F_o = \sigma_o \cdot A_o \tag{15}$$

In Figure 8, the shear force, F, is plotted *versus* the deformation, Δl, of the sample for 3 polybutylacrylates with different amounts of the chain transfer agent DMCT, as indicated. The test area of the samples is 12.7×12.7 mm^2, *i.e.* l_o is 12.7 mm. The sample with 0.5% DMCT (curve a) which is a material with a comparatively low viscosity ($\eta_o = 3 \cdot 10^3$ Pa·s) shows a linear decrease of F with increasing Δl. The measured F *versus* Δl curve is compared with the calculated straight line, showing a fairly good correlation. The PBA with 0.05% DMCT (b), which is a polymer near the gel point, exhibits a different shear curve with a pronounced maximum and a steep decrease of F followed by a slow decrease to the failure point at l_o. The deviation from a linear decrease is still more pronounced for the PBA without DMCT (c). This slightly crosslinked polymer shows a high force maximum and cohesive failure already at an elongation of the sample of about 1 mm. It is obvious that the dynamic shear behaviour of the second and third sample cannot be modeled by the viscosity-based approach, shown above. We come, thus, to a similar conclusion as in the case of the static shear test, namely that polymers with sufficiently low viscosities which exhibit a linear decrease of F follow the predictions from linear viscoelasticity, whilst highly viscous and crosslinked materials show significant deviations from this approach with a high force maximum and a separation at small elongations.

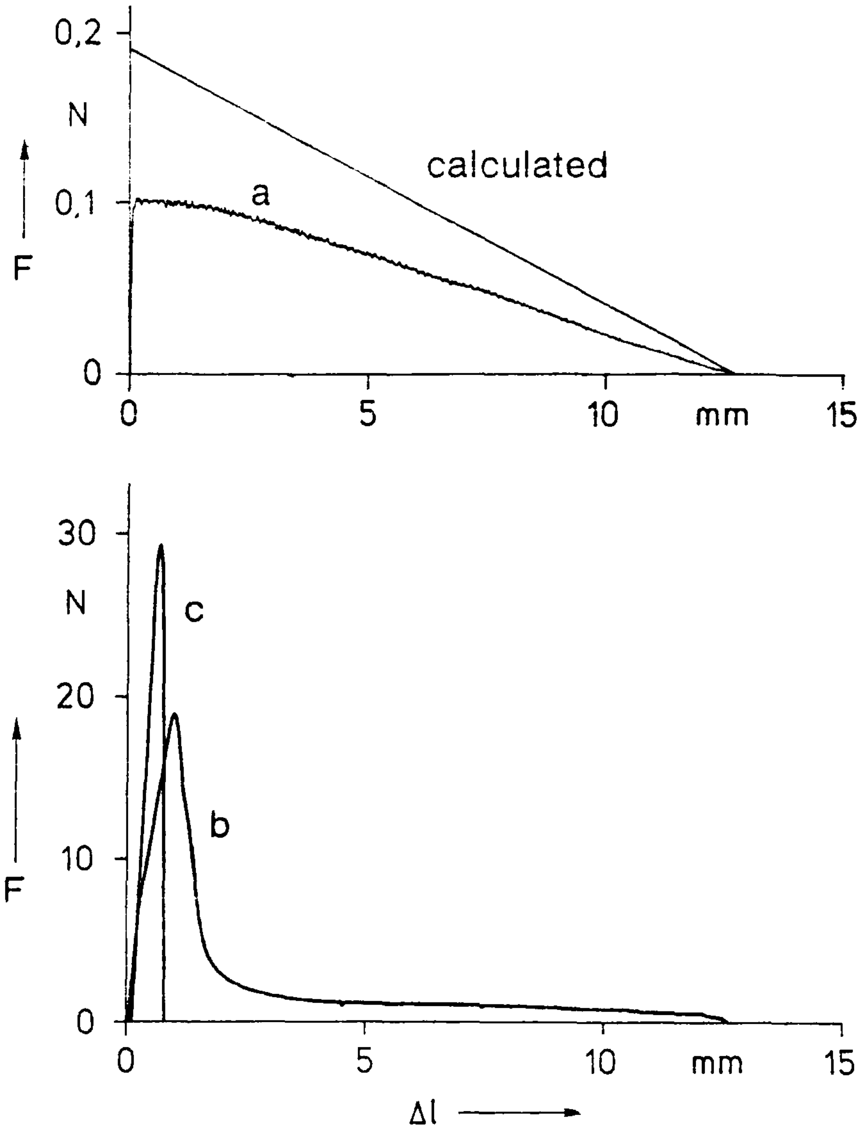


FIGURE 8 Force *versus* elongation Δl curves for dynamic shear tests on polybutylacrylate samples with different concentrations, c, of t-dodecylmercaptane. T = 23°C. a: c = 0.5%, straight line calculated according to equations (13) to (15), b: c = 0.05%, c: c = 0%.

In order to establish an empirical relation between the dynamic shear strength and viscoelastic data which can be applied to a larger range of polymers, we try to correlate the measured shear force, F_o, or the corresponding shear stress, σ_o, which is defined by equation (15), to the shear stress, σ_o, calculated according to equation (13), which can be written as

$$\sigma_o = |\eta^*| \cdot \omega_d = |G^*(\omega_d)| \,. \qquad (16)$$

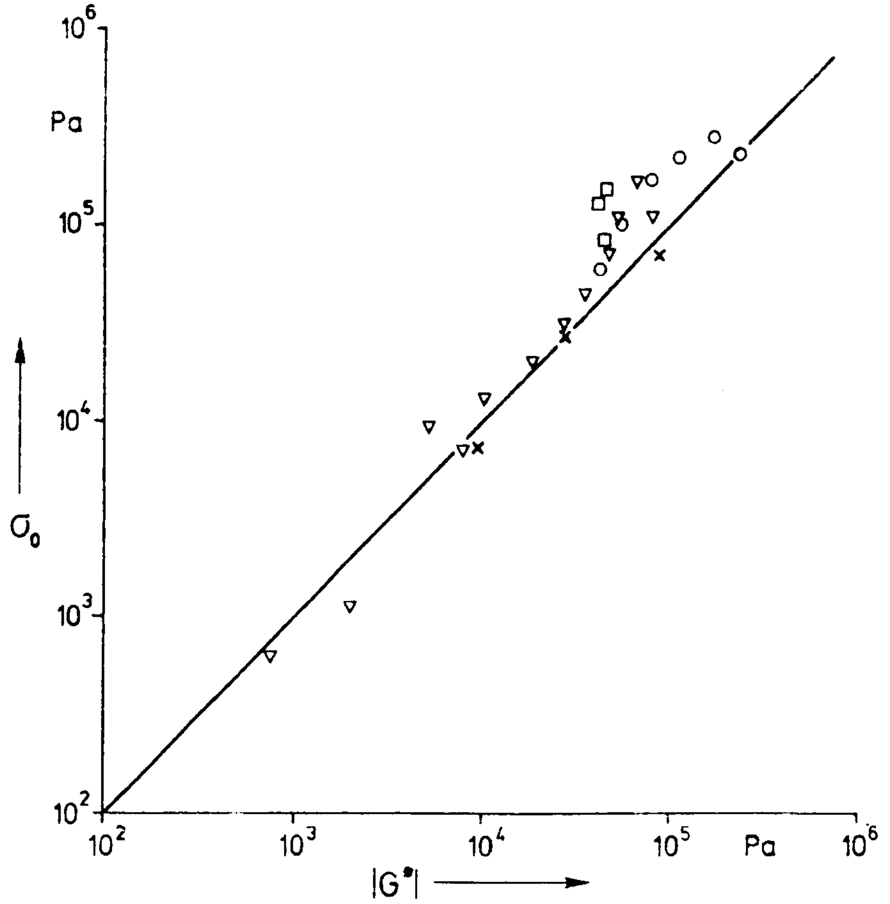


FIGURE 9 Dynamic shear strength (maximum shear stress σ_o), plotted *versus* the dynamic shear modulus, $|G^*|$, at the angular frequency, ω_d, equal to the shear rate of the dynamic test. Samples: Polybutylacrylate with DMCT (▽), BDA2 (□) and MAA (○), polyisobutylene (×).

In Figure 9, the measured shear stress, σ_o, is plotted *versus* the absolute value of the shear modulus, $|G^*|$, determined at the angular frequency, ω_d, equal to the shear rate, $\dot{\gamma}_d$, of the dynamic shear test. As expected, the agreement is good in the range of moduli and shear stresses, σ_o, below about $3 \cdot 10^4$ Pa, which equals a viscosity of about $1 \cdot 10^4$ Pa·s, as the dynamic tests were carried out with a shear rate of 3.3 s^{-1}. At higher moduli, the measured shear stresses tend to be higher than the corresponding moduli. That means that an exact correlation is not found for polymers with high viscosities or even crosslinking.

5. CONCLUSIONS

The stress strain histories of both shear tests, static as well as dynamic, have been studied in this work. For polymers with low or moderate viscosities, the shear strain *vs.* time characteristics in a static shear test and, accordingly, the static shear strength, the holding time, t_c, can be calculated from the master curves of the dynamic shear modulus. The range in which this agreement is valid is not defined very sharply but is found for zero shear viscosities below about $1 \cdot 10^6$ Pa·s.

For the dynamic shear test, the dependence of the shear force on the sample deformation or the corresponding shear strain can be calculated, as well. This approach is restricted to samples which have a viscosity at the shear rate of the dynamic test below about $1 \cdot 10^4$ Pa·s.

These exact calculations cannot be applied to highly viscous or slightly crosslinked polymers which, however, are the materials used in the pressure sensitive adhesive industry. On the basis of the model experiments, empirical correlations between the shear strength and the dynamic shear modulus could be established which seem to be valid more generally. For the static shear test, an average shear rate is calculated from the holding time. The static shear data in this study confirm that the shear modulus, $|G^*|$, determined at the angular frequency equal to this shear rate, falls into a relatively narrow range.

This empirical correlation surely does not mean that the nonlinear mechanical behaviour of highly viscous or crosslinked polymers in a static shear test is determined by linear viscoelasticity. It is simply an indication that the polymers studied here are in a somewhat similar viscoelastic state at the time scale of the static shear test, which seems reasonable.

In the case of the dynamic shear test, a relation is found between the dynamic shear strength (σ_0) and the shear modulus, $|G^*|$, at the angular frequency equal to the shear rate in the test.

The correlations shown in Figures 7 and 9 are not exact enough to allow the substitution of the nowadays-applied shear tests by mechanical measurements, such as the determination of the dynamic shear modulus, and to calculate the shear strength from the moduli. Dynamic mechanical measurements can, at the utmost, be used for a rough estimate of the shear strength. The significance of these relations should be seen more in the potential to understand better both shear tests and to correlate shear data to the molecular structure of pressure sensitive adhesives, which is reflected by the shape of the modulus *versus* frequency curves.

Acknowledgements

Appreciation and thanks are given to Dr. Kröner for the polymerization of the model polymers, to Dr. Dittrich for the standard shear measurements, and to Dr. Dittrich and Mr. Müller for many helpful discussions and for their assistance in preparing the manuscript.

References

1. PSTC-7 "Holding Power of Pressure-Sensitive Tape" (revised 1985).
2. AFERA Test Method 4012 "Measurement of shear adhesion" (1979).
3. FINAT Test Method 8 (FTM8) "Resistance to shear from a standard surface" (1985).
4. J. Johnston, *Adhesives Age* **32** (No. 12), 30 (1990).
5. A. Zosel, Proc. PSTC First World Congress for Pressure Sensitive Adhesive Tape (Pressure Sensitive Tape Council, Skokie, IL, 1992), p. 191.
6. Z. Miyagi and K. Yamamoto, *J. Adhesion* **21,** 243 (1987).
7. J. D. Ferry, *Viscoelastic Properties of Polymers*, 3rd ed. (J. Wiley, New York, 1980).

8. S. G. Chu in L.-H. Lee, Ed., *Adhesive Bonding* (Plenum Press, New York, 1991), p. 97.
9. W. P. Cox and E. H. Merz, *J. Polym. Sci.* **28,** 619 (1958).
10. C. A. Dahlquist in R. L. Patrick, Ed., *Adhesion and Adhesives*, Vol. 2 (Marcel Dekker, New York, 1969), p. 219.
11. N. A. deBruyne, *Nature* **189,** 262 (1957).
12. M. A. Krenceski and J. F. Johnson, *Polym. Engn. Sci.* **29,** 36 (1989).
13. W. C. Dale, M. D. Paster and J. K. Haynes, *J. Adhesion* **31,** 1 (1989).
14. E. H. Andrews, T. A. Khan, P. Drew and R. Rance, *J. Appl. Polym. Sci.* **41,** 595 (1990).