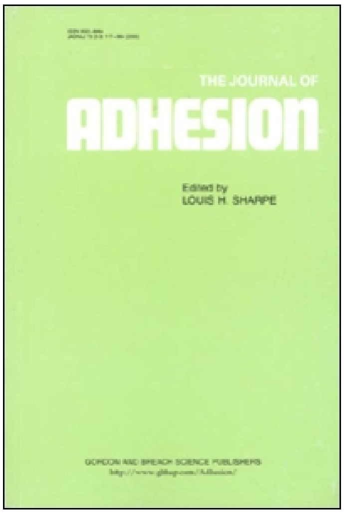

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

Viscoelastic Properties of Pressure-Sensitive Adhesives

E. P. Chang[a]
[a] Avery Research Center, Pasadena, CA, USA

To cite this Article Chang, E. P.(1997) 'Viscoelastic Properties of Pressure-Sensitive Adhesives', The Journal of Adhesion, 60: 1, 233 — 248
To link to this Article: DOI: 10.1080/00218469708014421
URL: http://dx.doi.org/10.1080/00218469708014421

J. Adhesion, 1997, Vol. 60, pp. 233–248
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Viscoelastic Properties of Pressure-Sensitive Adhesives*

E. P. CHANG

Avery Research Center, Pasadena, CA 91107, USA

(Received April 19, 1995; in final form December 14, 1995)

Recent studies on the correlation of viscoelastic properties of pressure-sensitive adhesive (PSAs) with industry standard performances such as peel, tack and shear are reviewed and discussed. One notewothy feature in these correlations is the separation of the bonding and debonding steps in PSA adhesion, which specifies the characteristic bonding and debonding frequencies of different PSA tests. Viscoelastic windows (VW) for different types of pressure-sensitive adhesives (PSAs) proposed by these workers are also compared and discussed. The observed good correlations reaffirm the importance of bulk viscoelastic properties to PSA adhesion performances.

KEY WORDS: Pressure-sensitive adhesives; dynamic mechanical analysis; viscoelastic properties, dynamic storage and loss moduli; bonding and debonding frequencies; peel; shear and tack performance; viscoelastic windows.

I. INTRODUCTION

It has been well established by many investigators that the performance of PSAs (*e.g.* peel, tack and shear) depends strongly on the bulk viscoelastic properties of the adhesives.[1–12] The William Landel Ferry (WLF) superposition procedure between rates and temperatures of the tests has been applied very successfully in adhesion tests both in peel[2,3] and in other modes of debonding.[4,7,8] In addition, correlations of different peel failure modes with different rheological regions of PSAs have been established and reported by Aubrey and Sherriff.[10] Dale and coworkers,[13] by combining the small-strain (dynamic mechanical) and high strain (stress-strain) measurements, and correlating these mechanical properties with industry standard "applications" peel and shear properties, established that the majority of the performance range shown by commerical PSA's is controlled by the bulk mechanical properties (tensile strength, storage modulus and dissipation) of the adhesive polymers. In addition, room temperature performance properties were found to correlate better with properties measured by DMA at higher temperatures than those at room temperature, suggesting the trade-off of high strain and high temperature. Recently, Tse[14] has identified the correspondence of adhesive performance frequencies with adhesive deformation frequencies on the rheological master curves. He proposed that the criteria for good PSAs are low plateau modulus (satisfying the Dahlquist contact criterion), and high-energy

*Presented at the Eighteenth Annual Meeting of The Adhesion Society, Inc., Hilton Head Island, South Carolina, U.S.A., February 19–22, 1995.

dissipation at the corresponding debonding frequency. Chu,[15] by comparing the dynamic mechanical properties of commonly employed elastomers and resins, together with their blends, and showing how they can be related to PSA industry standard test methods, was able to establish that the performance of commerical PSAs can be related to the glass temperature (T_g) and plateau modulus as well as the the frequency dependence of dynamic testing. Based on this principle, a viscoleastic window for good PSAs was proposed. More recently, viscoleastic windows of different types of PSA's based on dynamic storage (G′) and loss (G″) moduli at bonding and debonding frequencies have also been proposed by Chang.[16] The objective of this paper is to review the use of viscoelastic properties for the correlation of pressure sensitive adhesive performance in these recent works.

II. DISCUSSION

II.a. Review of Work of Dale *et al.*[13,28]

Even though the time scales of the mechanical experiments were not close to those of either peel or shear, they nevertheless presented some useful correlations between the mechanical results and the standard peel and shear data. This work identifies the superposition of high-temperature, small strain, dynamic mechanical measurements with room temperature, high-strain, peel and shear measurements. Figures 1 and 2 show, respectively, the excellent correlations between 20-minute peel strength *versus* log tan δ (dynamic mechanical loss factor) at 127°C, and log shear failure time *versus* 127°C storage modulus. They also recognized the decisive dissipation in peel is at high

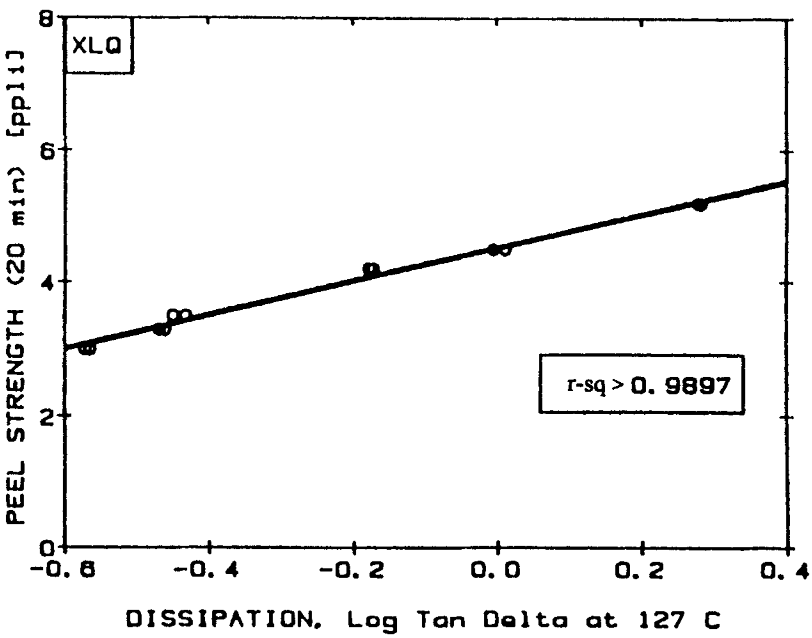

FIGURE 1 20 miniute peel strength versus log tan δ at 127°C for the crosslinker series. (Reprinted from Ref. 13 by kind permission of Gordon and Breach Science Publishers S.A.).

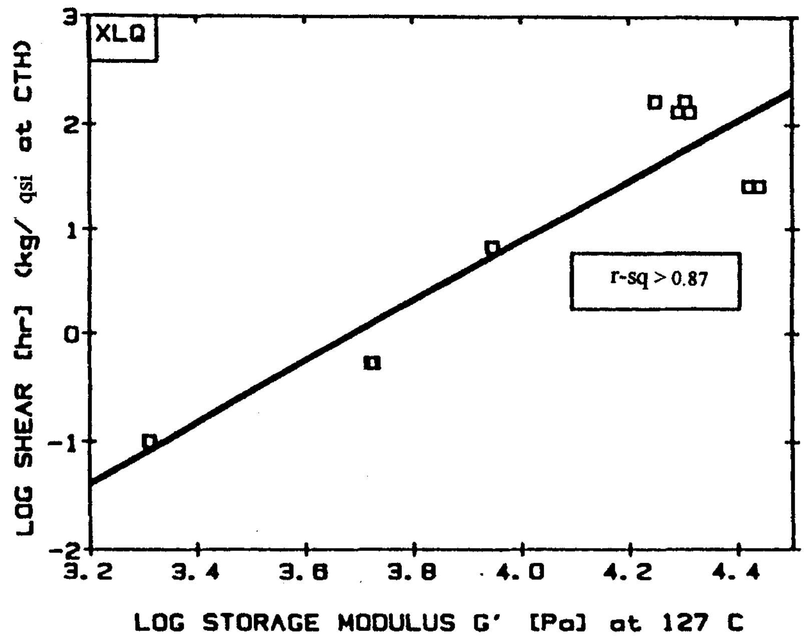

FIGURE 2 Log of shear hang time *versus* log storage modulus at 127°C for the crosslinker series. (Reprinted from Ref. 13 by kind permission of Gordon and Breach Science Publishers S.A.).

strains (closer to the tensile tests), and the caution of using tan δ as a measure of energy dissipation because of its strain dependency.[17,18]

The noteworthy features and contribution of this work are:

- The proposed strain temperature superposition concept to reconcile or correlate the small deformation dynamic mechanical and the large deformation tensile tests.
- The demonstration that the necessary requirements of successful PSAs are to possess simultaneously both solid-like strength and liquid-like flow behavior.
- The use of log tan δ (positive and negative, respectively) to delineate liquid-like and solid-like properties, and the caution of the strain dependence of tan δ.
- The importance of the differences in energy integral for adhesive (lower boundary stress) and cohesive peel failures (utilising full stress-strain curve) adhesives. This identifies the possibility of achieving higher peel strength going from adhesive to cohesive failure, *e.g.* on substrates of increasingly greater surface energy. This also confirms that the surface adhesion serves primarily to prevent premature separation from the substrate[2,19-22] that results in a low peel force.
- Confirmation of the major role that bulk viscoelastic properties play in PSA performance, and demonstration of the power of the tensile and dynamic mechanical methods for correlations with standard peel and shear performance tests.

II.b. Review of Work of Tse[14,29]

By combining the theoretical consideration of Kinloch and coworkers,[23-25] the Dahlquist contact criterion[1] and the proposed bonding and debonding functions, Tse presented the PSA performance as:

$$T = P_0 BD$$

Where T is the adhesion performance and P_0 is an intrinsic interfacial failure energy (either the energy required to open up a unit area of PSA-substrate interface in the absence of viscoelastic energy loss or the thermodynamic work of adhesion which is substrate dependent). B is the bonding function, assumed to be constant when the Dahlquist contact criterion is satisfied (*i.e* the plateau modulus is lower than 3.3×10^5 Pascals). D is the debonding function which is the viscoelastic loss component. It is strongly dependent on the characteristic debonding frequency, or the separation speed of the PSA test.

Even though viscoelastic measurements involve low strains, while PSA adhesion tests involve high strains, his study confirms many findings of earlier studies on the good correlations of viscoelastic properties with PSA adhesion. The noteworthy features and contributions of this work are:

- Separation of the bonding and debonding steps in PSA adhesion.
- Relationship between viscoelastic behavior at different frequencies and PSA performance and the identification and location of the debonding frequencies for different adhesion tests on the rheological master curves (Table I and Fig. 3).
- The proposal of a domain temperature, T_{dd}, which is much lower than the domain disappearance (critical) temperature, T_c, proposed by Krause and Hashimoto[26] and the "monophasic" temperature by Widmaier and Meyer.[27] The absence of correlation between shear adhesion failure temperature (SAFT) and T_{dd}, suggests that the responsible mechanism for the lower SAFT observed in resin-rich systems is the lowering of the plateau modulus or narrowing of the plateau width.
- Evidence both for and against the existence of two phases (polyisoprene-rich and resin-rich) in the midblock rubber matrix. Such differences can most probably be reconciled by the different sensitivities and thermal history of the different tests (DSC *versus* rheological measurements).
- The inference that resin restricts segmental motion of the rubber midblock, resulting in a higher monomeric friction coefficient.
- The identified criteria for good PSAs, namely, low plateau modulus to facilitate bonding and high energy dissipation at the PSA debonding frequencies and domain integrity. This is consistent with the proposed viscoelastic windows by Chu[15] and Chang.[16]

II.c. Review of Work of Chu[15,30]

Even though there had been earlier studies on the subject of dynamic mechanical properties of resin-rubber mixtures, Chu, nevertheless, through his systematic studies, presented a coherent picture of how the interaction of rubber with resins affects the dynamic mechanical properties, which is turn affects PSA performance. Compared with previous publications, the noteworthy features and contribution of these papers are:

- Identification of the limitation and strengths/capabilities of different Rheometrics fixtures (torsional rectangular, and different diameter parallel plates) for dynamic

TABLE I
Charactristic Debonding Frequency of the PSA Test (From reference 14)

Standard PSA Test	Test Frequency $= \frac{2\pi \text{ (bond rupture speed)}}{\text{adhesive thickness}}$ (rad/s)
Loop	140
Peel	435
Quick Stick	870
Probe	1650

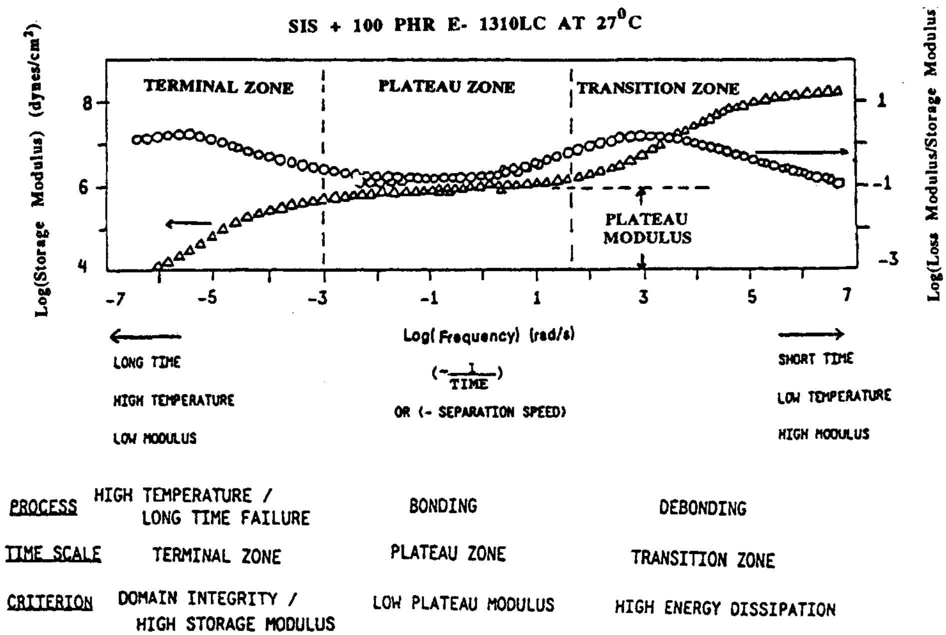

FIGURE 3 Relationship between viscoelastic behavior at different frequencies and PSA performance. (Reprinted from Ref. 14 by kind permission of the publishers, VSP).

mechanical measurements of PSA materials. Of particular values to those not too familiar with all the testing models, are the caution of instrument compliance at low temperatures or measuring glassy modulus and the recommendation of different size parallel plates for different modulus range measurements.

- The testing and chacteristics of resin-rubber compatibility by the criteria of a pronounced shift in the tan δ peak maximum temperature together with a decrease in the plateau modulus.
- The observation that rubbers with higher plateau modulus and T_g values are much more difficult to tackify than those with lower values. This criterion should compliment the matching of the solubility parameter between rubber and resin, commonly used in the PSA industries.
- The order of compatibility of different types of resins with different types of rubbers. This is particularly useful for formulation chemists who need guidance on the selection of compatible resins for these rubbers.

- The identification that good PSA systems (rubber tackified with appropriate amount of compatible resin) have a depression of modulus at low frequencies (making bonding favorable) and an elevation in the high frequency modulus (making debonding stronger). This also demonstrates the importance of $G'_{\omega=100}/G'_{\omega=0.1}$ in achieving good PSA properties (Fig. 4).
- The proposal of room temperature modulus values at different frequencies for PSA systems, *e.g.* for tapes and labels (Fig. 5), and the proposal of a viscoelastic window for good PSA tapes based on modulus requirement (Fig. 6).
- The correlation of tack data *versus* dynamic mechanical data, specifically $G'_{\omega=0.1}$ and ratio of $F'_{\omega=100}/G'_{\omega=0.1}$. The correlation points out that low $G'_{\omega=0.1}$ and high $G'_{\omega=100}/G'_{\omega=0.1}$ ratio are desirable for high tack values.
- The proposal of empirical windows for good PSA and good labels based on the loci of room temperature plot of modulus and T_g values determined from the temperature at which tan δ is a maximum (Fig. 7).

II.d. Work of Chang[16]

Using 10^{-2} and 10^2 rad/s as the frequency window for the peel, tack and shear tests, G′ and G″ values of different PSA samples at these two frequencies are measured, and their viscoelastic windows (VWs) were constructed by plotting the four coordinates: (1) G′ at 10^{-2} rad/s, G″ at 10^{-2} rad/s, (2) G′ at 10^2 rad/s, G″ at 10^{-2} rad/s, (3) G′ at 10^{-2} rad/s, G″ at 10^2 rad/s, and (4) G′ at 10^2 rad/s, G″ at 10^2 rad/s on the log-log cross plot of G′

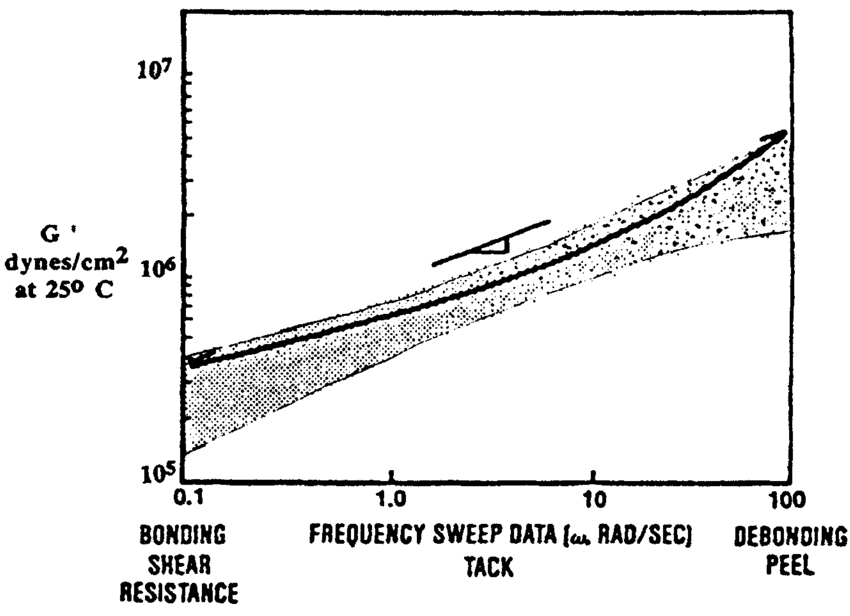

FIGURE 4 Room-temperature modulus requirement at various frequencies. Shaded area is the window which has good properties. The slope ($G'_{\omega=100}/G'_{\omega=0.1}$) is an important factor. (Reprinted from Ref. 15 by kind permission of the publisher, Plenum Press).

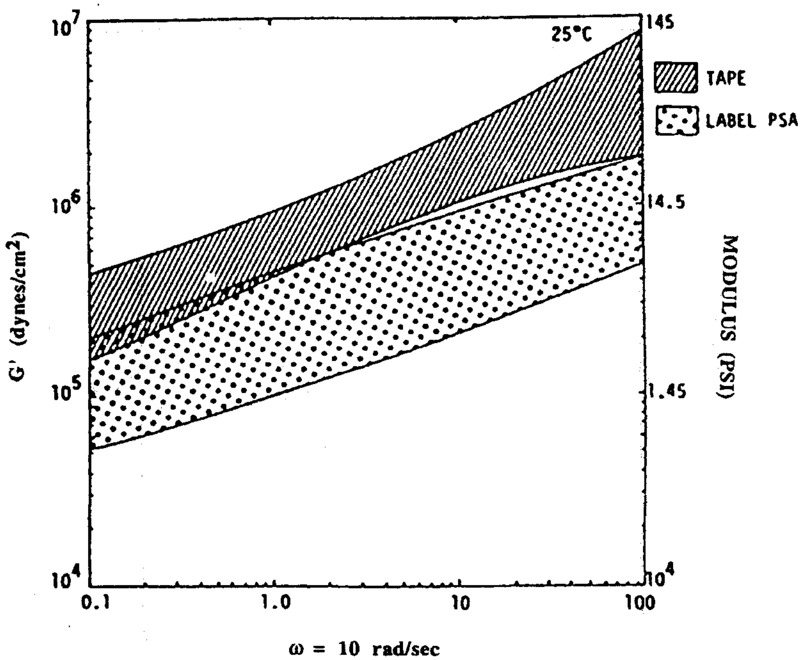

FIGURE 5 Room-temperature modulus values of label and PSA tape at various frequencies. (Reprinted from Ref. 15 by kind permission of the publisher, Plenum Press).

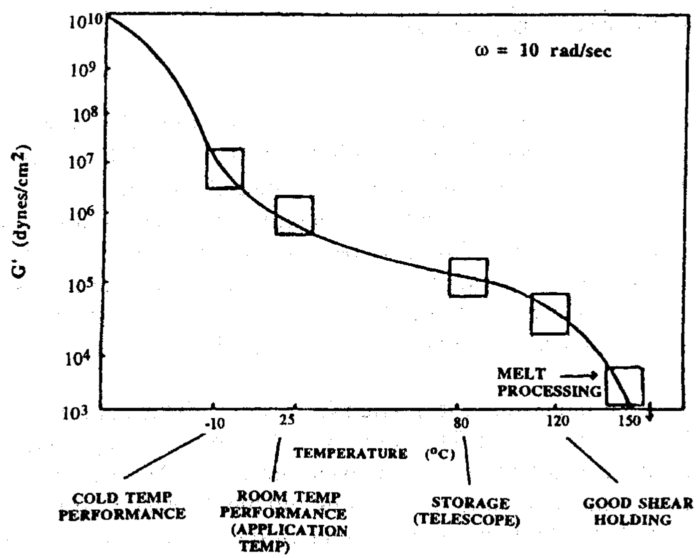

FIGURE 6 Modulus (G') requirements (windows) for good PSA tapes (Reprinted from Ref. 15 by kind permission of the publisher, Plenum Press).

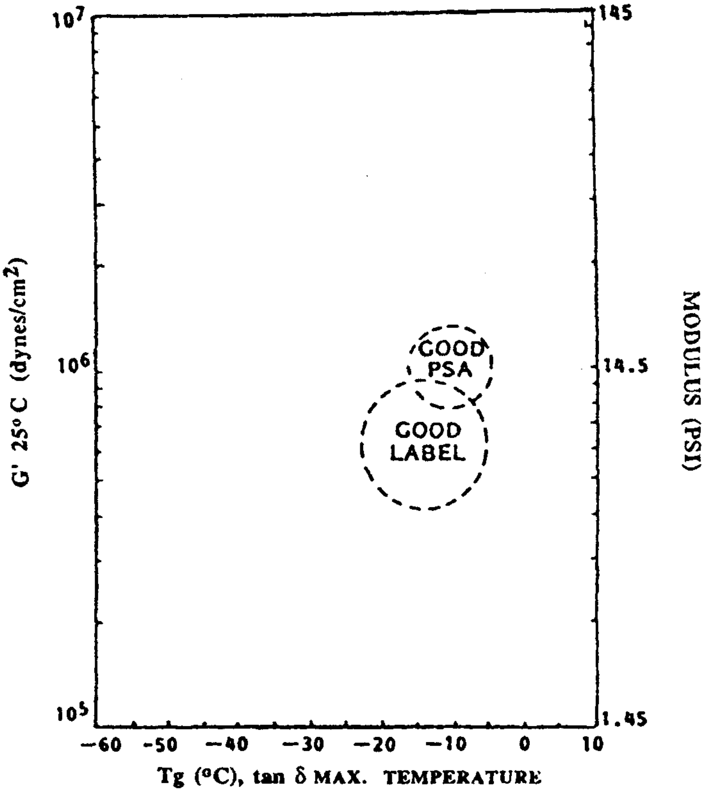

FIGURE 7 Empirical windows required for good PSA and lables (Modified and Reprinted from Ref. 15 by kind permission of the publisher, Plenum Press).

and G″. It was found that for most PSAs, the range of G′ and G″ at room temperature within the selected frequencies falls between 10^3 and 10^6 Pascals. In addition, there is a unique correlation between the adhesion performance of the PSAs *versus* the location of their VWs. A four-quadrant concept was, therefore, adopted to categorize different types of PSAs. The location of different viscoelastic windows is shown in Figure 8 with their corresponding operative viscoelastic regions :To illustrate the consistency of the viscoelastic window concept for the different types of PSAs illustrated, several key materials in each group are shown below to ascertain their viscoelastic uniqueness.

Quadrant 1: (Top left hand quadrant): High G′-low G″—This quadrant corresponds to high modulus, low dissipation. The bonding and debonding frequencies both occur at the plateau region of the rheological master curve. No PSA can be found in this quadrant beacuse of the high bonding modulus and highly elastic nature (lack of flow) of the material making the bonding step unfavorable. Some elastomer and release coatings occupy this quadrant.

Figure 9 shows the viscoelastic windows of a polydimethyl siloxane (PDMS), PDMS + 40% control-release additive (CRA), and PDMS + 60%CRA. It can be

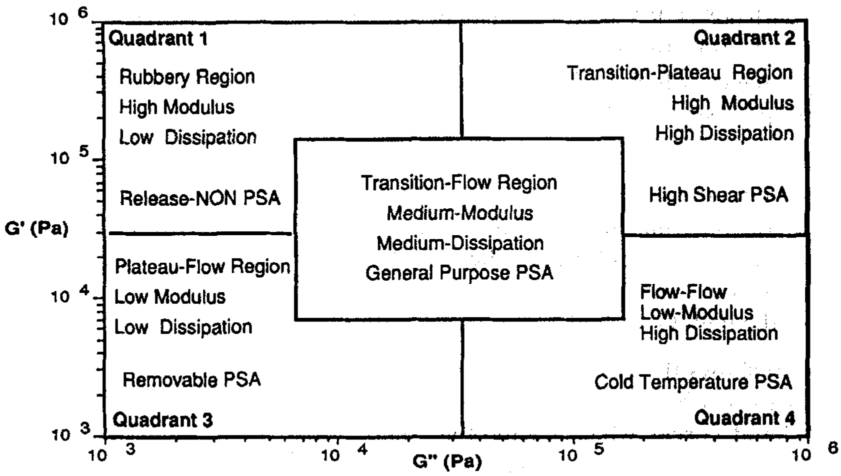

FIGURE 8 Viscoelastic windows of PSAs as related to different regions on their rheological master curves. (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

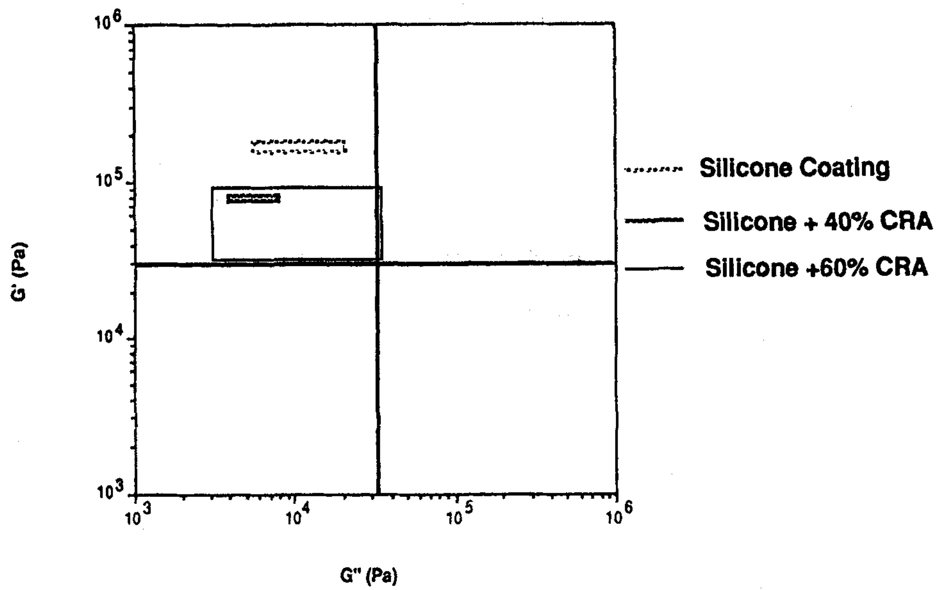

FIGURE 9 Viscoelastic windows of release coatings (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

observed that because of the high modulus (G′) and low dissipation (G″), PDMS is not a PSA, but rather is a release coating. However, with the incorporation of an increasing amount of CRA, the modulus is progressively reduced together with a progressive increase of flow or dissipation. Such progressive decrease in G′ and increase in G″ results in a progressive increase in the tackiness of the samples. Thus, PDMS, when modified with 60% CRA, is tacky. It can be anticipated that with further increase in the CRA concentration, the viscoelastic window of the resulting sample will move towards the central region which is the location for general purpose PSAs or, in this case, a general purpose silicone PSA.

Quadrant 2: (Top right hand quadrant): High G′-high G″—This quadrant corresponds to high modulus and high dissipation. The bonding frequency corresponds to the plateau region, while the debonding frequency corresponds to the transition in their rheological master curves for high shear PSAs. The high bonding modulus compensated by the high dissipation or flow makes the bonding marginal. Shear is high because of the high G′ or high cohesive strength of the material.

Figure 10 shows the viscoelastic windows of three high-shear PSAs: HSPSA-1, 2, and 3. It can be observed that all these adhesives' viscoelastic windows occupy the top-right hand corner which means that these adhesives have comparatively high modulus (G′) and high dissipation (G″) within the application rates (*i.e.* between 10^{-2} to 10^{2} sec^{-1}). In general these adhesives have comparatively high T_gs and are comparatively highly crosslinked to achieve the high shear performance.

Quadrant 3: (Bottom left hand quadrant): Low G′-low G″—This quadrant corresponds to low modulus, low dissipation. The bonding frequency corresponds to the plateau region, while the debonding frequency corresponds to the onset of the flow transition in their rheological master curves for removable PSAs. Bonding is facilitated by the low modulus in spite of the low flow characteristics. Peel values are usually low

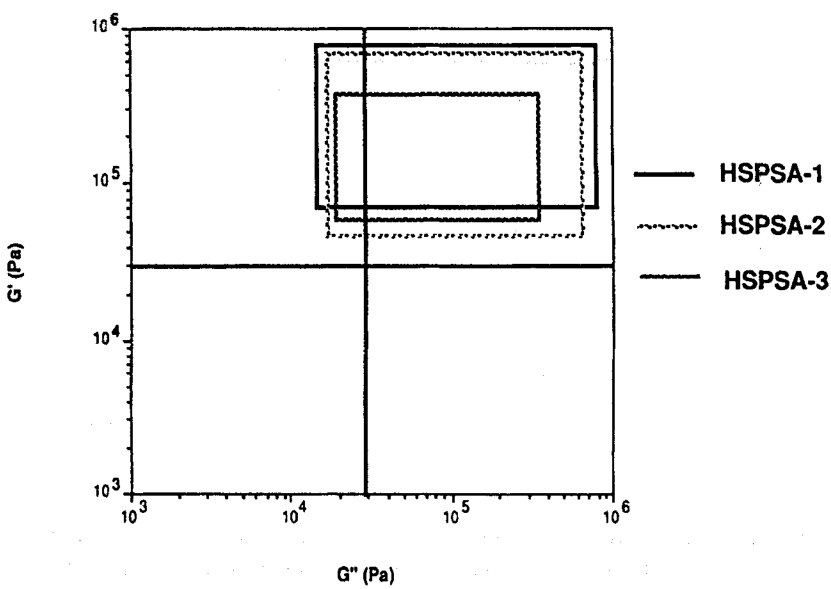

FIGURE 10 Viscoelastic windows of High Shear PSAs (Reprinted from Ref. 16 by kind permission of Gordon and Breach Publishers S.A.).

because of the comparatively low debonding strength and low dissipation. Removable and medical type PSAs fall within this quadrant.

Figure 11 shows the corresponding viscoelastic windows for removable adhesives RPSA-1, 2 and 3. The distinct characteristics of this type of adhesives are:

* Low bonding modulus so that the adhesive is very contact efficient.
* Low dissipation which implies more elasticity or better removability.

Figure 12 shows the viscoelastic windows of some of the removable PSAs used in medical applications (*e.g.* bandages). Comparing the viscoelastic windows of these medical removables with those removables in Figure 9, one notes that they tend to occupy the lower (better conformability) and further to the right (better flow) area of Quadrant 3. Some of the noteable differences between the removable and the bandage adhesives are:

* The reference temperature for the medical adhesive is the body temperature 37°C instead of 23°C in the removable case. This makes the bonding modulus of the medical adhesives even lower (more conformable) than the removables because of the higher reference temperature. This is desirable for contact area considerations because of the rough and frequently contaminated nature of the skin.
* The debonding moduli (top right hand corner of the window) are usually higher than those of the removables. This again is necessary to prevent lift or detachment because of frequent movement of the skin especially on curved areas such as knee and elbow.

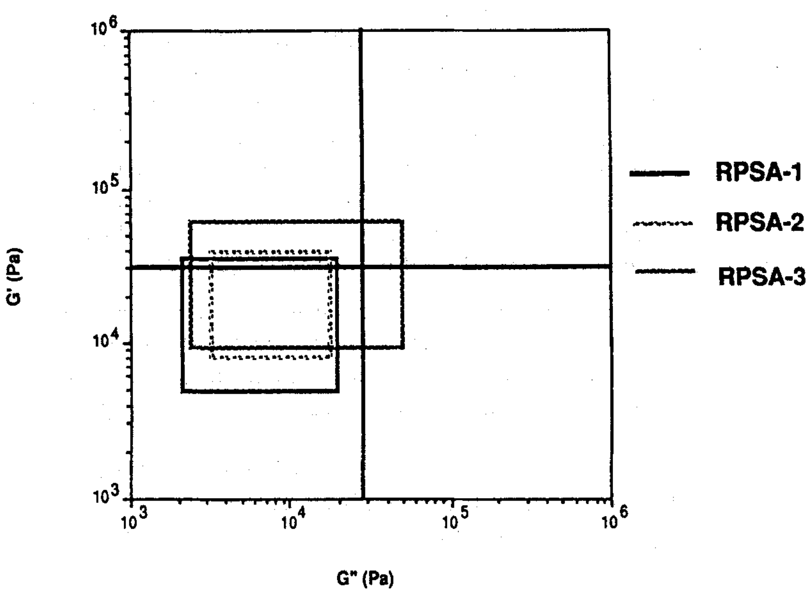

FIGURE 11 Viscoelastic windows of Removable PSAs (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

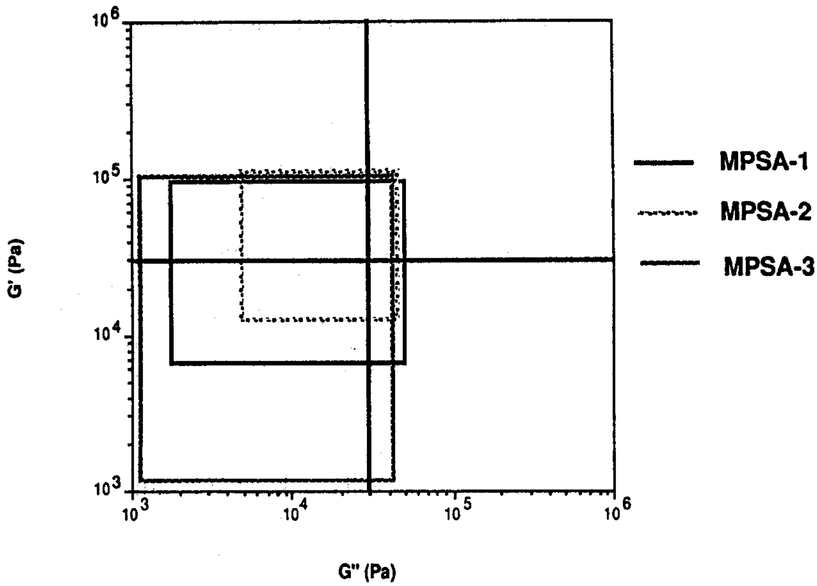

FIGURE 12 Viscoelastic windows of PSAs (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

Quadrant 4: (Bottom right hand quadrant): Low G′-high G″—This quadrant corresponds to low modulus-high dissipation . The bonding frequency corresponds to the onset of flow region, while the debonding frequency corresponds to the transition in their rheological master curves for very quick or cold-stick PSAs. The low bonding modulus coupled with high flow makes bonding very efficient, thus permitting the material to stick even at low temperature or very short contact time.

Three adhesives that have a significant portion of their viscoelastic windows into the 4th quadrant are (1) an emulsion all-temperature adhesive, ATPSA-1, a solvent cold-temperature adhesive, CTPSA-1, and a silicone PSA. Their windows, which indicate low bonding modulus as well as good flow nature, fall somewhere between the 3rd and the 4th quadrants (Fig. 13). So far, no good example of a PSA has been found with its viscoelastic window located right in the fourth quadrant.

Central Area: Medium G′-Medium G″—This central area corresponds to medium modulus-medium dissipation. The bonding frequency corresponds to the transition region, while the debonding frequency corresponds to the flow region in the rheological master curves (usually characterized by the absence of a distinct plateau region) of general purpose PSAs.

Figure 14 shows the corresponding viscoelastic windows for three general-purpose acrylic PSAs, GPPSA-1, 2 and 3. It can be observed that they all occupy the central region (overlapping part of the four quadrants) illustrating the general-purpose nature of this type of PSA.

Correlation and Prediction of Adhesive Performance with the Viscoelastic Window

Figure 15 illustrates the relative position of the viscoelastic window with respect to the

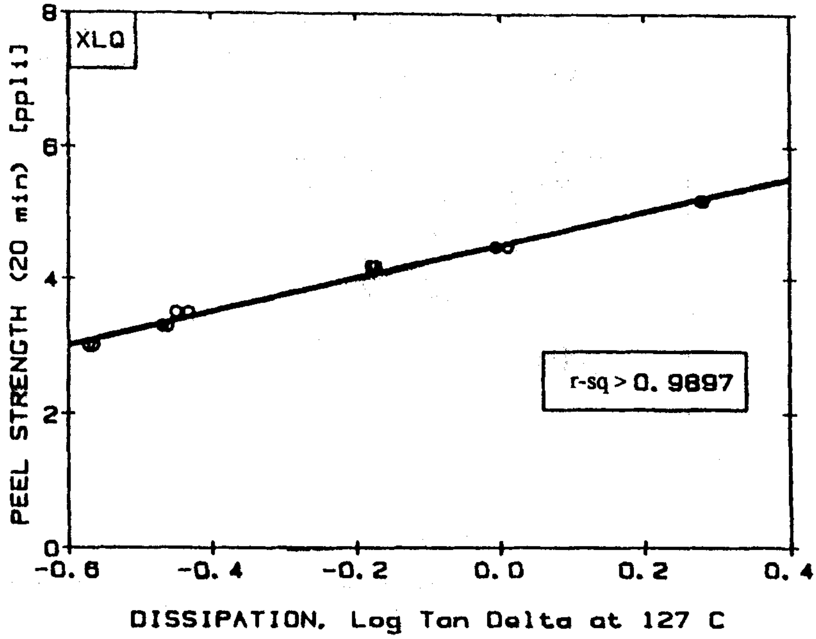

FIGURE 13 Viscoelastic windows of selected all-temperature and cold temperature PSAs (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

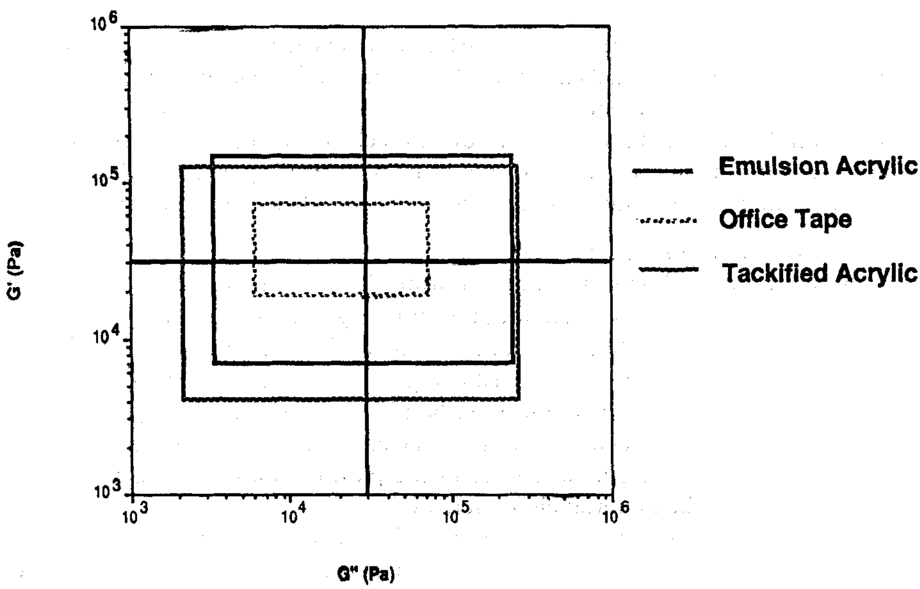

FIGURE 14 Viscoelastic windows of general purpose PSAs (Reprinted from Ref. 16 by kind permission of Gordon and Breach Science Publishers S.A.).

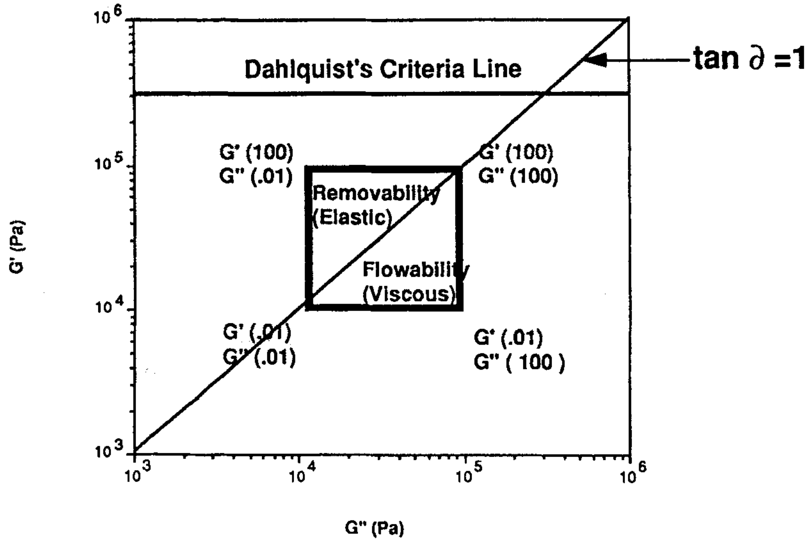

FIGURE 15 Relationship of the viscoelastic window with the Dahlquist contact criteria and tan δ (Re-printed from Ref. 16 by kind permission of Grodon and Breach Science Publishers S.A.).

Dahlquist Contact Criteria line[1] as well as the diagonal line where $G' = G''$ or $\tan\delta = 1$.

The Dahlquist Contact Criteria Line

The Dahlquist line is an important reference line as it indicates whether a material would be contact efficient (PSA) or deficient (non-PSA).[1] It is quite evident that except for the release coatings, all the different types of the adhesives shown in Figures 9 to 14 have a bonding modulus (*i.e.* the base of the window) much below the Dahlquist line (which means good conformability). In other words, by comparing the position of the base of the window with the Dahlquist line, immediately we know whether the material is a PSA.

The G'-G" Cross-Over Line ($\tan\partial = 1$)

The diagonal $\tan\partial = 1$ line is another important line of demarcation as it separates regions where the elastic or storage modulus (G') is greater (*i.e.* $\tan\delta < 1$) or smaller ($\tan\delta > 1$) than the loss modulus, G". The portion of the window to the left of the line (*i.e.* $\tan\delta < 1$) indicates the more elastic region. In other words, the closer the viscoelastic window is towards the top-left hand corner of the G'-G" plot, the more elastic (or better removablity) will be the material characteristics. Conversly, the closer it is towards the lower right hand corner of the plot, the more viscous (or cohesive failure prone) will be the material characteristics.

Assuming that the adhesive is the only variable in the construction, and that the surface effect is negligible, the following adhesion and convertibility performance can be correlated with the shape and location of the viscoelastic window.

Shear Performance

The shear performance can be correlated with the following features of the window. The base of the window (*i.e.* G' at 0.01 rad/s.) usually indicates the value of the plateau modulus in view of the subambient T_g of PSAs. In general, the higher the plateau modulus (provided the Dahlquist Contact Criteria is still satisfied), the better will be the shear. The high shear type of adhesive is a good manifestation of this correlation. In addition, if the base of the window or the plateau modulus is the same, the more extended the plateau, (*i.e.* the difference between G′ at .01 and 100 rad/sec. is smaller), usually, the better is the shear. This is because a more extended or flatter rubbery plateau is indicative of either higher degree of entanglement due to higher molecular weight, higher chemical or physical crosslink density. Since the breadth of the plateau is inversely proportional to the height of the window, if the base of the window is the same, the shorter the window, the better will be the shear performance prediction.

Peel Performance

Peel performance is dependent upon the efficiency of the bonding step as well as the separation resistance in the debonding step. The bonding efficiency can be correlated with the plateau modulus at the bonding frequency (~ 0.01 rad/s). In other words, the lower the G′ value at 0.01 sec^{-1} (or the base of the window), the more favorable is the bonding. The debonding strength comes from two contributing terms, the cohesive strength which is indicated by the storage modulus, G′, and the energy of dissipation term which is indicated by the loss modulus, G″, both measured at the debonding frequency (~ 100 rad/s). Thus, the higher the debonding G′ and G″ values (*i.e.* the top right-hand corner of the window), the higher will be the debonding strength.

Tack Performance

The correlation of tack performance is similar to that of peel, except the bonding frequency for tack is about 1 rad/s, which means that the bonding efficiency relates approximately to the inverse of the half-height of the window. The debonding resistance can be related again to how high is the top-right hand corner of the window (*i.e.* G′ and G″ values at 100 rad/s)

The usefulness of Chang's viscoelastic window concept is:

(1) It can be obtained by making two measurements at two frequencies (*i.e.* 10^{-2} sec^{-1} and 10^2 sec^{-1} at the specified temperature). Such simple and rapid measurements will immediately identify the nature and type of the adhesive.
(2) With the viscoelastic window defined, qualitative information regarding adhesion performance and the mode of failure can be obtained as described. This is particularly useful for comparing, evaluating and screening different adhesives since the shape and location of their viscoelastic windows provide comparative qualitative performance information prior to measuring their peel, tack and shear performances.

The limitation of Chang's viscoelastic window method is that it only gives the G′ and G″ values at those two frequencies. There is no information for G′ and G″ at the in-between frequencies (e.g. 10^{-1}, 10^0, 10^1 sec^{-1}, etc.) or other frequencies as in either the full frequency scan between the viscoelastic window frequencies, or a master curve which would be the recommended method for more quantitative information.

III. SUMMARY

Recent studies on the correlation of viscoelastic properties of pressure-sensitive adhesives (PSAs) with industry standard performances such as peel, tack and shear have been reviewed and discussed. One noteworthy feature which improves the correlations is the separation of the bonding and debonding steps in PSA adhesion, which specifies the characteristic bonding and debonding frequencies of the different PSA tests. One common viscoelastic requirement for a good performance PSA from these studies is the low bonding plateau modulus at the bonding frequency and high-energy dissipation at the debonding frequency. This is reflected in the proposed viscoelastic windows (VW) for different types of pressure-sensitive adhesives (PSAs).

Acknowledgements

The author would like to thank Avery Dennision for permission to publish this work.

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