

**Society for Adhesion and Adhesives**

**and**

**Applied Polymer Science Committee**

**PRESSURE SENSITIVE  
ADHESIVES**

**One-day Symposium 25<sup>th</sup> April, 2001**

**Society of Chemical Industry, Belgrave Square,  
London**

## **Programme**

- 10.00 Registration and coffee**
- 10.30 Measuring adhesive tack**  
B Duncan, (NPL Teddington)
- 11.00 Fundamentals and structure property relationships**  
A Zosel, (BASF Germany)
- 11.30 Adhesion of soft adhesives: a microscopic view**  
C Creton, (PCSM,-ESPCI France)
- 12.00 The effect of flexible substrates on PSA performance**  
A Steven-Fountain, (Reading University)
- 12.30 Lunch**
- 14.15 Using molecular modelling as a practical aid to adhesive design**  
D Porter, (DERA Farnborough)
- 14.45 Polymer resins for waterborne pressure-sensitive adhesives: Challenges for industrial R & D**  
O Dupont and S van Es, (UCB Belgium)
- 15.15 PSA polymerisation – effects of process change**  
T Jones, (Harlow Chemical Co.)
- 15.45 Waterborne pressure-sensitive adhesives; correlation of properties with particle structures**  
P Lovell, (UMIST Manchester)
- 16.15 Tea**

This one-day symposium is one of an on going series organised by the Society for Adhesion and Adhesives.

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## Loop Tack Measurements

Bruce Duncan, NPL Materials Centre, Teddington, Middlesex, TW11 0LW

### Introduction

The capability to rapidly and reliably bond two surfaces is a key performance requirement in many pressure sensitive adhesive products, particularly adhesive tapes and labels. A key property for successful bonding is the tack or 'stickiness' of the adhesive. Tacky materials can be discerned by touch but for material specification or product development purposes a more quantitative approach is necessary. Tack is defined as the ability of an adhesive to form a bond to a surface after brief contact under light pressure. Often thought of as a simple property to measure, tack depends on a complex interaction of many different factors and, hence, there is no single value that characterises tack.

#### *Tack is a complex property*

##### *Bond formation depends on:*

- Adherend surface properties - material, wettability or surface energy, roughness, porosity.
- Preparation - cleanliness, pre-treatments, coating weight and uniformity, adhesive application, open or drying time, environmental conditions experienced prior to bonding.
- Physical and Chemical properties of the adhesive - type, functional groups, flow properties, surface energy,
- Bonding process - contact pressure, duration of contact, rate of pressure change, thermal history, penetration into surface.

##### *Separation is influenced by:*

- Separation process - rate of separation, angle of peel, specimen clamping.
- Mechanical properties of adherends - flexibility, modulus, cohesive strength of surface layers.
- Mechanical properties of adhesive - rigidity, cohesive strength, extension to failure, visco-elastic properties, creep, stress relaxation.

##### *Determined values also depend on:*

- Quantities measured - separation force or amount of peel., sampling rate.

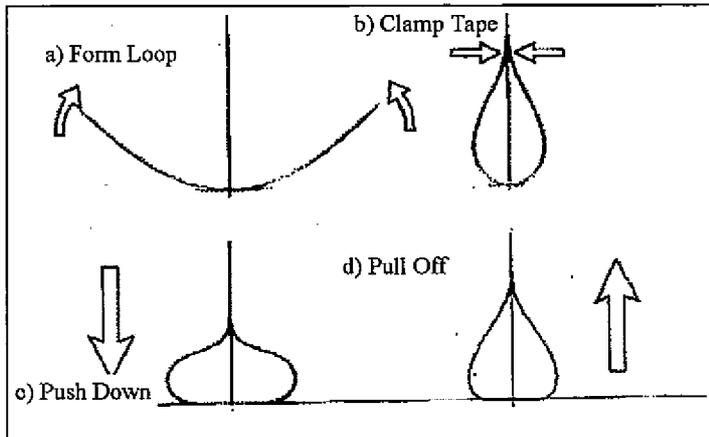
Many measurement methods for tack have been developed by industries for specific applications and even within a sector of industry there can be a myriad of different measurement methods in use. Standard measurement methods for pressure sensitive adhesive tapes include loop tack, probe tack and rolling ball tests. It is often the case that measured tack values depend very strongly on the test method [1]. There is often little correlation, even for qualitative ranking, between different measurement methods. Poor reproducibility between measurement laboratories is common despite well-specified tests. This was illustrated in a round-robin study where, despite being supplied with a detailed test procedure, nearly a third of the participating laboratories were unable to correctly rank three tapes having significantly different levels of tack. This makes specification and control of tack properties difficult.

Whatever the adhesive application or industry sector, there are common factors affecting tack and similar measurement processes used - bonds must be made and broken in order to assess the strength of the bond. In collaboration with organisations in the packaging industry (Pira International) and footwear Industry (SATRA), the National Physical Laboratory has been taking a more in depth look into the measurement of tack.

Pressure sensitive adhesives are visco-elastic polymers whose mechanical properties are strongly dependent on temperature and strain rates. In theory, the temperature of tests can be controlled so that differences between methods can be eliminated. However, the rate sensitivity of the adhesive's properties manifests itself in both the spreading of the adhesive to form intimate contact with the surface during bonding and the behaviour during separation. Loading rates and contact times differ between test methods. Hence, the visco-elastic behaviour of the adhesives is a key factor to the poor agreement

between different test methods. Understanding this and choosing a method where the stress history approximates that in the bonding application is important to ensure that measured tack properties are relevant.

### Loop Tack Tests



Loop tack tests are intended for quality control and specification of adhesive tapes and pressure sensitive adhesives. Standard methods usually specify the length and width (generally 25 mm) of the loop strip, the dimensions and material of the base plate and the speed of the test. Tack is defined as the force required to separate, at a specified speed, a loop that has adhesively contacted a specified area of defined surfaces. The tests can be performed with the adhesive coating either the tape or the base.

**Figure 1:** Steps in the Loop Tack Test

Each standard method specifies surface, cleaning and sample preparation steps. Once this is done all types of loop tack test can be performed in the same way. There are four steps in the test, Figure 1.

*Step a:* Form the loop from the tape.

The specified length of tape should be bent back, with the adhesive outermost, until around 10 mm or so of the ends are in contact. Tapes with extremely low stiffness can be difficult to handle (particularly if static electricity causes the backings to stick together). To avoid such problems the tape could be bent around a cylinder or tube of appropriate diameter when forming the loop. The ends of the tape should be joined (using adhesive tape) to ease clamping. Alternatively, short strips of metal can be bonded to the ends of the tape to reduce problems with handling or clamping.

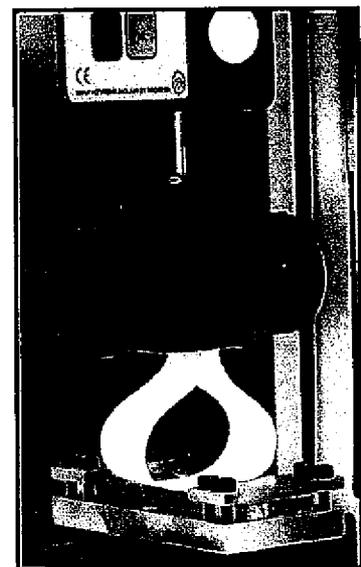
*Step b:* Clamp the loop in the movable test machine grips.

The grips should be connected to a load measuring device with sufficient range and sensitivity (normally specified in the test method). The loop should be aligned such that the edges of the tape will be at a right angle to the edge of the base plate.

*Step c:* Lower the loop.

The loop should be lowered, pushing down onto the base surface, until the tape contacts over the required area which is normally the whole width of the base. Determination of full contact can be subjective (Figure 2). Although the standard test methods do not require it, measurement of the 'push down force' can help check for consistency amongst different tapes. The exact shape of the loop, and thus the distribution of stress, is determined by the stiffness of the tape.

*Step d:* Pull the loop off the surface

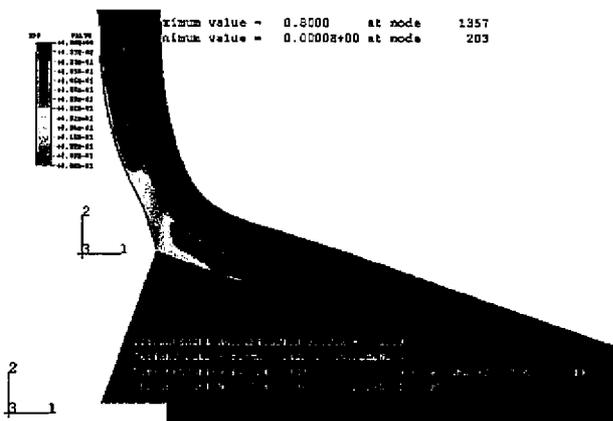


**Figure 2:** A loop tack test

Once the loop has contacted the required area of the base plate the direction of the test machine is reversed. The area in contact is inspected visually for any imperfections in the contact (e.g. wrinkles or bubbles) and the results omitted from the analysis should this occur. The test runs until the tape is detached from the plate. Typically, the force may show one or more peaks before final separation. Some methods [2, 3] specify that tack is taken from the peak force whilst others [4] are more ambiguous. The de-bonding of the loop is a dynamic process and response rate of the force transducer or logging device can influence the maximum recorded. Low response rate devices will tend to miss the peak force and record slightly lower tack than high response devices. A study indicated that there may be 5 % difference between the maximum forces determined using fast (1200 points per second) and slow (10 points per second) devices [5]. The standard methods tend to specify different ways of expressing the tack. A single force value for tack may obscure the more complex behaviour of the system. The height of the peak is one parameter of the tack but the failure energy is also represented in the breadth of the peak.

### Effects of Substrates

Loop tack tests, as with all adhesive joint tests, measure the aggregate response of the bonded system. Therefore, the mechanical responses of both the adhesive and bonded parts influence the measured tack



values. Differences in the mechanical properties (e.g. modulus, thickness and clamping) of the backing and/or the base can give significantly different tack values despite identical surfaces, adhesives and processing conditions. This effect of adherend stiffness on the nominal 'tack' can be explained by the changes in the forces required to deform the bonded surfaces and the different stress states in the adhesive layers due to changing angles of peel. Such effects can be analysed by Finite Element mechanics and Figure 3 contrasts the predicted peel (at similar peel forces) of a flexible backing, rigid base with that for a stiffer rigid backing, compliant base case. Where the base is compliant there is a large deformation of the base. This significantly changes the stress and strain distribution in the adhesive layer by increasing shear strain but reducing principle strain in the adhesive. Therefore, at the same peel force, adhesive rupture is much more likely in the case of the compliant backing, rigid base than the case where the base is more compliant.

Rigid base, low stiffness backing.

Compliant base, stiffer backing

Figure 3: Predicted strain distributions in peel tests

Some effects of the influence of backing stiffness and base compliance are shown, in Figure 4, through plots of maximum principle strain in the adhesive layer against peel force. These show that there can be large differences in peel forces required to achieve a consistent level of strain. If this strain is a rupture criterion for the adhesive then the difference sets of mechanical properties for the backing and base will lead to significantly different tack values.

Materials properties data and reliable models to characterise the deformation and failure of flexible visco-elastic adhesives under complex states of stress are lacking. Normal measurement techniques for visco-elastic properties, such as dynamic mechanical analysis or oscillatory rheometry, operate predominantly in the low strain region whereas tack strength is also determined by the large strain, failure behaviour of the adhesive. Methods for determining the mechanical performance of flexible adhesives are the subject of a current research programme.

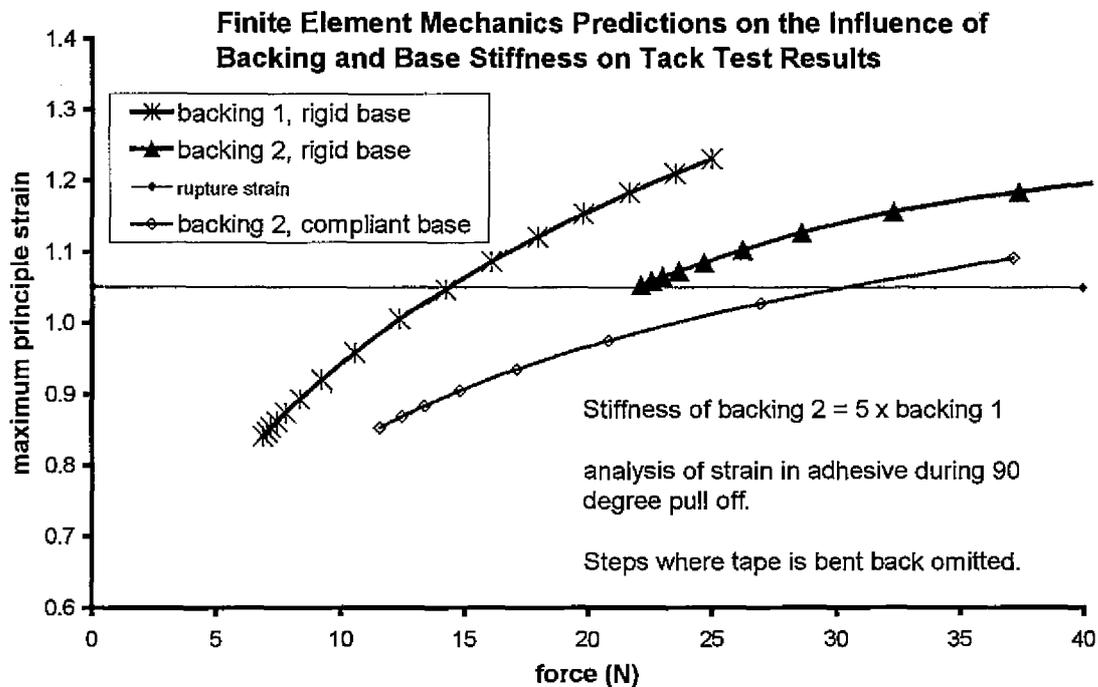


Figure 4: Results of FEA model of loop tack tests

### Conclusion

In conclusion, loop tack tests are often seen as simple tools for quality control. However, interpreting and correlating these tests is complicated by the visco-elastic nature of the adhesive and the significant influences of the mechanical properties of the components bonded. Improved materials properties data and mechanical modelling tools will enable a better understanding of this 'simple' test method.

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# FUNDAMENTALS AND STRUCTURE PROPERTY RELATIONSHIPS

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## 1. Introduction, adhesion performance of PSA's

As Pressure Sensitive Adhesives are used above their glass transition temperature, they have to be regarded in the larger context of the adhesion science of soft polymers, which experienced considerable progress during the last decades [1]. This paper presents correlations between the adhesion performance and the mechanical behaviour of PSA's on two levels: firstly between adhesion and the linear viscoelastic properties which are characterized by the components of the dynamic shear modulus, i.e. the storage and the loss modulus, measured with a dynamic shear rheometer with a parallel plate geometry. On a second stage, micromechanical deformation processes are discussed which cannot be deduced from linear viscoelasticity and which give rise to high energy dissipation.

The adhesion performance of PSA's is primarily characterized by three quantities, tack, peel strength and shear resistance, for which a number of standardized test methods is available [2]. Tack and peel strength show a relatively similar behaviour with respect to the time and rate scales of the debonding processes. The shear strength describes the behaviour of PSA's under the influence of long lasting stresses. It is normally determined with creep measurements (in a first approximation) on a much longer time scale and often shows an opposite dependence on molecular parameters as the two other quantities, i. e. an increase in tack or peel strength is connected with a decrease in shear strength and vice versa.

## 2. Adhesive failure energy and tack

The adhesion performance of PSA's, especially the tack, is governed by the conditions of bond formation as well as bond separation. A major experimental progress has been achieved by methods which control the main factors influencing bonding and debonding, i.e. contact pressure and time, rate of separation and temperature, and determine the adhesive failure energy. A flat ended probe is brought into contact with the surface of the adhesive, and the force versus time curve of bonding and debonding, the maximum force of separation and the adhesive failure energy per unit of interface are determined [3]. The tack is defined as the adhesive failure energy, determined under conditions of low contact pressure and short contact time.

Measurements of the dynamic shear modulus and the adhesive failure energy of a number of polymers in dependence on temperature show that the tack has a maximum in the temperature interval above the glass transition range where the viscoelastic behaviour is governed by entanglements [4] (Figure 1). This gives a first indication that entanglements are a major molecular parameter controlling adhesion and tack of PSA's. Dynamic mechanical analysis has proven to be a very useful

instrument for studying structure property relations in PSA's, e.g. giving insight into the influence of entanglement length, crosslink density, and the role of tackifying resins [4].

Two types of stress versus strain curves have been observed in our investigations of a large number of polymers [5] (Figure 2). Type a, observed for polymers with low tack, shows a sharp stress maximum and adhesive debonding at comparatively low strains, i.e. some similarity with brittle deformation behaviour. Type b with a stress maximum and an extended plateau, leading to a large area under the curve and a high strain at break, is observed for polymers with high tack. It has some similarity with the stress strain characteristics of tough polymers with yielding or plastic deformation.

It was found by high speed photography and video optic observation of the debonding process in probe tack experiments that debonding starts with the formation of cavities in the polymer or the polymer probe interface, presumably at preexisting nuclei, which is caused by the high dilatant stresses at the beginning of the deformation process [3,6]. We have found cavitation to occur in all polymers, studied so far. For polymers with low tack the cavities remain small after nucleation, and the tensile stress increases rapidly until failure occurs in the interface. In polymers with low plateau modulus, i. e. high entanglement length, the cavities grow very rapidly after nucleation until they form some kind of foam like or fibrillar structure. The micro-mechanics of bond separation is strongly affected by the temperature and the rate of separation [7].

The molecular mass between entanglements, the so-called entanglement length, has a strong effect on fibrillation or lamellation. Polymers with an entanglement length above about  $1$  to  $1.5 \cdot 10^4$  g/mole show fibrillation whilst materials with an entanglement length below this limit debond by homogeneous deformation.

### 3. Peel strength

It has been observed rather early that the peel force, measured in dependence on peel rate, is related to Young's modulus, plotted versus the rate of elongation or the reciprocal angular frequency [8]. A transition in the peel strength at low peel rates, often connected with a transition between cohesive and interfacial failure, is associated with the transition from liquidlike to rubberlike behaviour of the adhesive, whilst the peel transition at high rates corresponds to the transition from rubberlike to glassy behaviour. At this transition, very often the so-called stick-slip behaviour is observed which makes its appearance in periodic oscillations of the peel force and in chaotic, jerky motions of the contact line.

Kaelble seems to be the first who observed that the peeling edge becomes unstable and splits up into fingers penetrating into the contact area [9]. Impressive pictures of deformation patterns during peeling, i.e. fibrillar or lamellar structures, have been published by Urahama [10] and Verdier et al [11]. That means that the micromechanics of the separation processes is very similar in probe tack and peel experiments respectively, taking into account the different homogeneity or heterogeneity of the stress fields.

#### 4. Shear resistance

The shear strength of a PSA is characterized by the time, required to pull a defined area of an adhesive tape from a test panel under a constant load. Investigations with an instrument, which simulates this test and measures the shear strain with time under a constant applied shear force, show that the shear strain versus time curves can be calculated from the dynamic shear viscosity for samples with comparatively low viscosities [12]. The shear versus time curves of highly viscous or slightly cross-linked polymers, however, which better represent the materials used as PSA's, show two parts: a slow increase of the strain with time over a long interval and a very steep, "catastrophic" increase of the strain until failure occurs (Figure 3). This shear characteristics cannot be calculated from viscoelastic data.

In order to get insight into the micromechanics of shear experiments we recently started video optic observations of the adhesive layer through a transparent test panel during the shear test. At the beginning of the "catastrophic" phase, failure starts within the adhesive layer at the edge of the lap joint, and at last forms a narrow band. This is followed by the rapid formation of further, parallel bands of cohesive failure. That means that there is a very inhomogeneous micromechanical deformation process present in shear experiments, too, which gives rise to the rapid break down at the end of the shear experiment. More work is needed in order to understand this micromechanics of the shear test.

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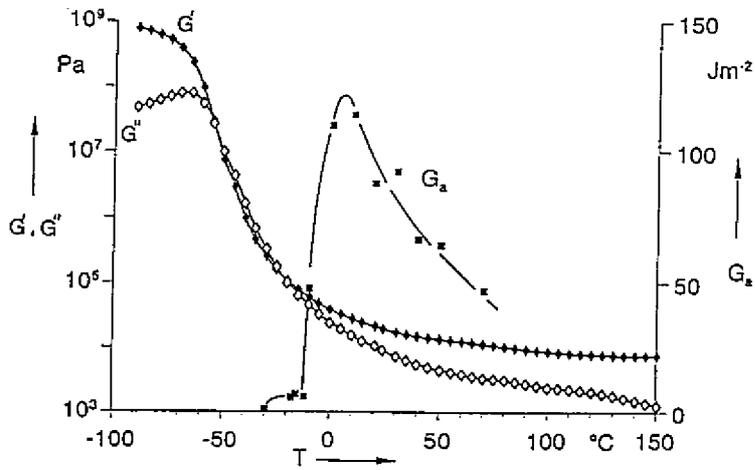


Fig. 1: Dynamic shear modulus  $G'$ ,  $G''$  and tack  $G_a$  in dependence on temperature for polyethylhexylacrylate

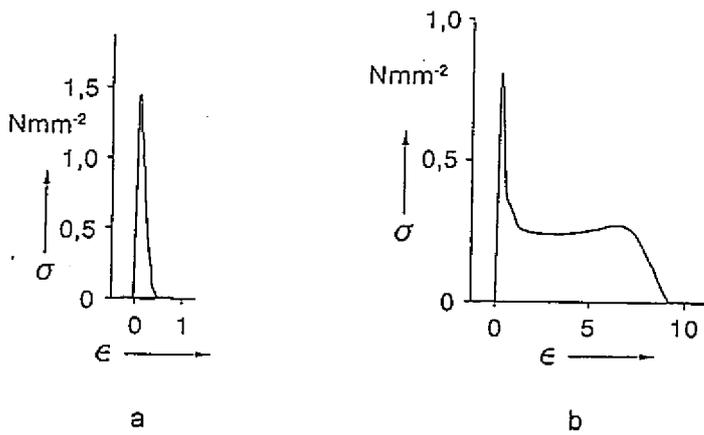


Fig. 2: Stress strain diagrams of the debonding process for a polymer with low tack (a) and high tack (b)

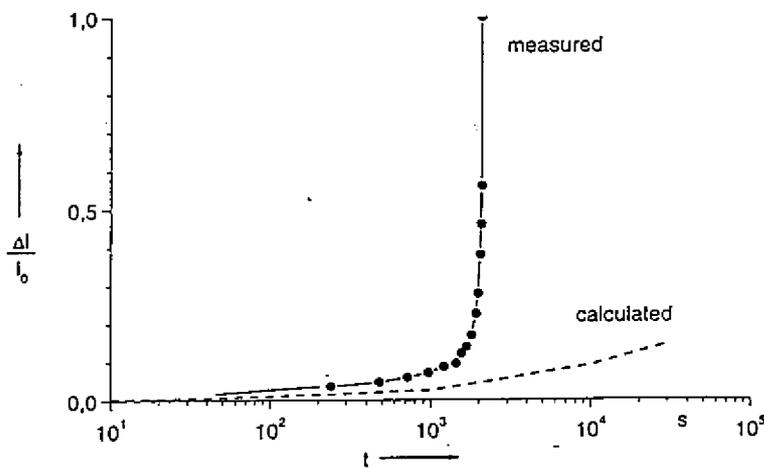


Fig. 3: Comparison of the calculated and measured shear versus time curve for an acrylic PSA

# ADHESION OF SOFT ADHESIVES: A MICROSCOPIC VIEW

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## I. INTRODUCTION

A variety of adhesion tests serving different purposes have been developed in the PSA industry. The flat-punch test, also called probe tack test, is widely used to test the short-time, low-pressure adhesion. This test has the advantage of applying a fairly uniform displacement (uniform stress field and strain rate) to the adhesive film over the whole surface of the probe, and for those soft systems where failure involves the formation of cavities and fibrils<sup>1</sup>, it facilitates the analysis of the process. The maximum nominal stress  $\sigma_{\max}$ , the maximum nominal strain  $\varepsilon_{\max}$ , and the adhesion energy,  $W_{\text{adh}}$ , defined as the integral under the stress-strain curves, are the main relevant parameters to characterise the performances of a PSA, and provide clues for the practical design of the molecular structure of the polymeric material for a given application. However, for a more refined optimisation of the properties, it is helpful to understand what really happens when testing the adhesive film and how its molecular structure is involved. New developments in both instrumentation and interpretation of the results have provided insights in the debonding mechanisms of soft adhesive layers. In particular the breakdown of the debonding process in separate stages allows a better understanding of the role of the molecular features of the adhesive and the coupling between the rheological properties of the adhesive and the surface properties of the adherent. Some of these new concepts are reviewed here with the help of experimental examples.

## II. INSTRUMENTED PROBE TACK TEST

We perform the mechanical measurement as following : the adhesive layer is deposited as a 50-100  $\mu\text{m}$  thick layer on a clean glass slide, and a flat cylindrical probe, of radius 3 to 5 mm, usually made of glass or stainless steel, is brought into contact with it and subsequently removed. If desired, the surface of the probe can be coated with a molecularly thin mineral or organic layer (soft/hard polymer...) in order to modify surface interactions. During the compression phase, we apply a nominal contact pressure of 1 MPa, in order to achieve a maximum contact area. The contact time can be varied between 1 sec and more than 1 hour, and the probe is then removed from the film at a constant probe velocity which can be varied between 1 and 1000  $\mu\text{m/s}$ . The temperature is controlled, and can be varied between  $-20^{\circ}\text{C}$  and  $60^{\circ}\text{C}$ . Our custom-designed probe tack apparatus allows the observation of the adhesive film from underneath the transparent substrate with a microscope and/or a video camera along with the simultaneous acquisition of a nominal stress and strain curves. The nominal stress  $\sigma$ , is defined as the force divided by the contact area, and the strain  $\varepsilon$ , is defined as the displacement divided by the thickness of the film. Details on the experimental setup can be found elsewhere<sup>2</sup>.

Video captures during the probe tack test of an acrylic PSA with a steel probe (high energy surface) show that the debonding process can be broken into separate stages:

1. Initiation of the failure process at low deformation through the formation of microscopic cavities at the interface between the film and the probe or in the bulk of the adhesive layer. The load-bearing area decreases, and as a direct consequence,  $\sigma$  goes through a maximum, defining  $\sigma_{\max}$ .
2. Expansion of these cavities, both in the lateral and vertical dimensions until, from the underneath, the structure looks like a foam.
3. Elongation of walls in between the cavities in the direction of the applied stress, at an approximately constant level of nominal stress, defining a plateau value,  $\sigma_{\text{plateau}}$ .
4. Fracture occurs by creeping (cohesive failure) or debonding of the foot of the polymer ligament from the probe, defining  $\varepsilon_{\max}$ .

For permanent PSA's, most of the total adhesion energy is dissipated during step 4, i.e. elongation of the polymer ligaments as walls between cavities and / or fibrils.

### III. MECHANICAL ANALYSIS AND EXPERIMENTAL RESULTS

Recent mechanical analyses of the flat-probe adhesion test of a soft viscoelastic polymer film have shed some light on the reasons for such behaviour<sup>3-5</sup>. The most common way for a sticky material to release the elastic strain energy when pulled off from a surface, where non covalent bonds act, is to propagate an interfacial crack. For soft adhesives, i.e. polymer melts or gels of low elastic modulus, this process is not favoured as it requires large bulk viscoelastic dissipations and the available strain elastic energy is not very large. Moreover, in the probe tack geometry, the lateral dimensions of the compliant adhesive layer, i.e. diameter of the probe, are significantly greater than its thickness,  $h$ . Consequently, as the layer is deformed in the thickness direction, lateral strains cannot be accommodated and significant lateral stresses develop. The resulting strong hydrostatic pressure induces an alternative deformation mode to the propagation of a crack, namely the cavitation process<sup>3</sup>.

#### Cavitation

Many years ago, it was noted by Gent<sup>6</sup> that a cross-linked elastomer could not sustain a hydrostatic stress higher than approximately its elastic modulus without undergoing cavitation, that is the unstable expansion of an existing defect into a macroscopic visible cavity. This mechanical analysis was extended to a polymer melt by considering its time dependent modulus<sup>7</sup>. For a polymer melt, the rapid growth of a cavity is predicted when :

$$\sigma_{\text{hyd}} > E(t) \quad (1)$$

where  $E(t)$  is the time dependant elastic modulus. In agreement with Kaelble predictions', and applying time-temperature superposition, we indeed observed a direct correlation between the shear elastic modulus  $G'(\omega)$  and the maximum stress  $\sigma_{\max}(V_{\text{deb}})$  for different model acrylic PSA's over a large range of frequency / debonding rates and temperatures<sup>2</sup>. An increase in  $\sigma_{\max}$  with an increasing rate of debonding or a decreasing temperature was observed, and that increase closely matched the increase of the elastic component of the shear modulus  $G'(\omega)$ .

Intuitively, both the adhesive and the probe surface roughness should impact the cavitation process, since air bubbles could be trapped at the interface<sup>8</sup> or an inhomogeneous strain field could be created from asperities on the surface. Both could act as nucleation sites for the formation of cavities. Such effects are observed for example, when the roughness of the probe is varied. Recent systematic experiments have shown that the amplitude of the surface asperities has a direct effect on the level of stress at which the cavities are formed<sup>9</sup>.

## Foam formation

Once a few cavities have appeared and as the adhesive film is further strained in the tensile direction, either the growth of existing cavities, or the appearance of new bubbles in between the existing ones can be observed. Following our mechanical argument, the nucleation of new cavities should stop when the reduced confinement of the remaining load-bearing area allows sufficient lateral strain accommodation. At this point, any further tensile displacement of the polymer film leads to two competing deformation mechanisms :

- Stretching in the tensile direction of the polymer ligament in between the cavities
- lateral propagation of cavities if the energy release rate  $G$  at the cavity edge is higher than the critical energy release rate  $G_c$ .

Substantial deformation in the polymer film occurs prior to failure only if the first situation is favoured. Otherwise, rapid detachment of the walls between cavities can be observed if the local critical energy release rate is rather low.

Using a spherical probe that reduces viscoelastic losses in the vicinity of the circular propagating crack, and a JKR type analysis, it was shown that the energy release rate  $G$  for acrylic elastomers could be adequately described by the following equation<sup>10</sup>:

$$G = G_o (1 + (v/v^*)^n) \quad (2)$$

$G_o$  is the value of the energy release rate at a vanishing crack velocity, which can sometimes be equated to the thermodynamic work of adhesion of the polymer melt on the surface, and the parameters  $v^*$  and  $n$  characterise the velocity dependence arising from viscoelastic dissipation in the vicinity of the crack tip. Although equation 2 cannot be applied directly in the confined geometry of a probe-tack test or for very viscoelastic materials, it gives an insight into the type of coupling existing between the rheological properties of the adhesive and the surface properties of the adherent.  $G_o$ , for typical PSA's on steel surfaces, is rather high and prevents extensive lateral crack propagation, favoring the foam formation and fibril stretching mechanism. Insight on the nature of the coupling can however be obtained from studies on model systems.

Substantial changes in  $G_o$  can be induced by varying the surface probe chemistry, for example when a steel surface is coated by a thin layer of poly(dimethyl siloxane) (PDMS). In this case,  $G_o$  is low on most PSA's and the cavities grow mainly at the interface and their coalescence lead to a rapid adhesive failure, or eventually, the cavitation process is suppressed for materials with a high elastic modulus<sup>11-13</sup>.

At the opposite, on a high energy surface, the energy release rate is mainly determined by the rheological properties of the adhesive. In the probe tack test geometry, for the same polymers, we observed a transition from a lateral growth dominated for a pure PnBA to an extensional growth dominated for a PnBA-AA<sup>14</sup>. In fact, for a given set of experimental parameters or applications, the relaxation times / rheological properties of the adhesive can be tuned by varying the molecular structure of its components, i.e. monomers polarity, cross-linking, chain branching, in order to favour the extensional growth regime.

## Elongation

The vertical elongation of the walls between cells implies that, at the molecular scale, there is a progressive orientation of the polymer chains in the direction of traction<sup>15,16</sup>. Some of the extensional growth is a result of a creeping process and some is due to the drawing of unoriented polymer from the film. The respective importance of the two mechanisms will depend on the rheological properties of the adhesive in elongation (large strain regime). For example, for a SIS based PSA, the ultimate failure occurs by detachment of the foot of the fibril from the surface of the adherent, while for a weakly strain hardening acrylic adhesive, some cohesive fracture of the fibrils could be observed<sup>2</sup>. The maximum extension achieved, and therefore the adhesion energy, depends on the balance between the elongational properties of the adhesive and the interfacial strength still characterised by  $G_0$ .

## V. CONCLUSION

The performance of PSA's is critically dependent on the fine control of their yielding behavior under relatively small stresses. This yielding behavior often occurs in highly confined geometries, and the ability to design tests with realistically large values of the confinement ratio is one of the advantages of the probe tack test. Moreover, because of the uniform strain and strain rate which are applied by a flat probe test, the debonding process can be easily broken down into more elementary steps and analyzed. As opposed to a more conventional test such as peel, an instrumented tack test provides information on the sensitivity of each specific step to the molecular features and experimental conditions. In particular we have been able to distinguish between the initiation of failure (controlled by the linear viscoelastic  $G'$ ), the interfacial propagation of a crack (controlled by  $G_c$ ) and the extension of a fibril (controlled by the nonlinear extensional properties of the adhesive).

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## The Effect of Flexible Substrates on PSA Performance

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### Abstract

Medical pressure-sensitive adhesives are designed for use on human skin, which is a highly non-linear, extremely compliant substrate. To improve wound dressings and related products, the mechanics of skin and the interactions between flexible substrates and pressure-sensitive adhesives need examining.

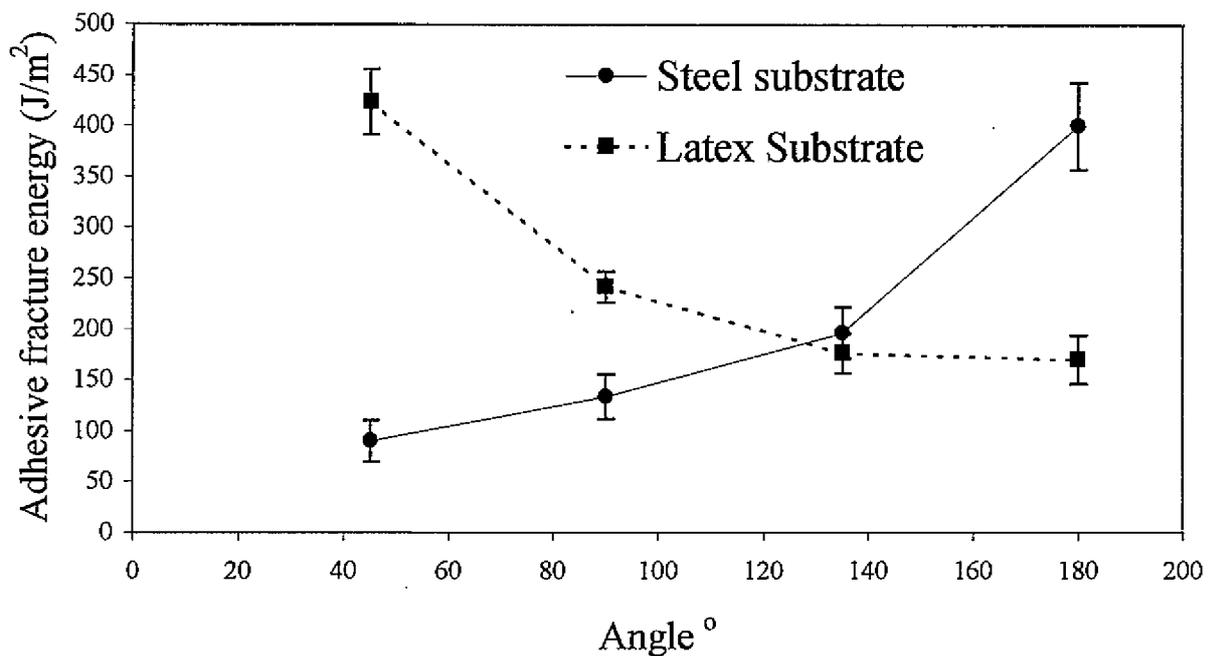
Many factors contribute to adhesive strength and removal of pressure-sensitive adhesives, including peel angle, peel rate, adhesive rheology and surface energy of the substrate. All these factors have been investigated traditionally using stiff substrates such as glass or steel. The geometry of peeling from a flexible substrate can be quite different from a rigid substrate. It has to be asked whether such differences in geometry affect peeling forces and the ranking of adhesives for “adhesion strength”.

Figure 1 shows peeling of 36 micron thick Melinex tape from steel and latex rubber substrates at different angles ( $\theta$ ). The adhesive fracture energy is determined from the peel forces using the well known relationship:

$$G = \frac{P}{b}(1 - \cos\theta) \quad (1)$$

When the peel forces from the flexible substrate are converted to  $G$  (still using Equation (1)), we see that different results are obtained. In particular, the influence of the peel angle is completely reversed. We believe that such results and come about because of the neglect of substrate flexibility.

Figure 1: Peel tests of Melinex tape from stiff and flexible substrates. Error bars represent standard deviation of five tests.



Different degrees of flexibility will also affect the peeling forces and give spurious  $G$  values when Equation (1) is used. For example, with neoprene rubber substrates, the peel forces for Melinex tape at 180° peel angle, for 1 mm and 2 mm thickness are very similar, Figure 2a. This is not really surprising as there is minimal deformation of the substrate in a 180° peel test. However, at a peel angle of 90° there is a clear difference between peeling forces for the different thicknesses, Figure 2b. The thinner substrate deforms more, the peel force is greater, and in consequence the  $G$  determined using Equation (1) is greater. The reason for these differences has to be due to the flexibility of the substrate. Furthermore, during a peel at a nominal angle of 90° the *local peel* angle changes throughout the test, and can be very different from the initial peel angle, used in Equation (1).

Figure 2a: Melinex peel test from neoprene rubber of different thickness, at a speed of 50mm/min., and nominal peel angle of 180°

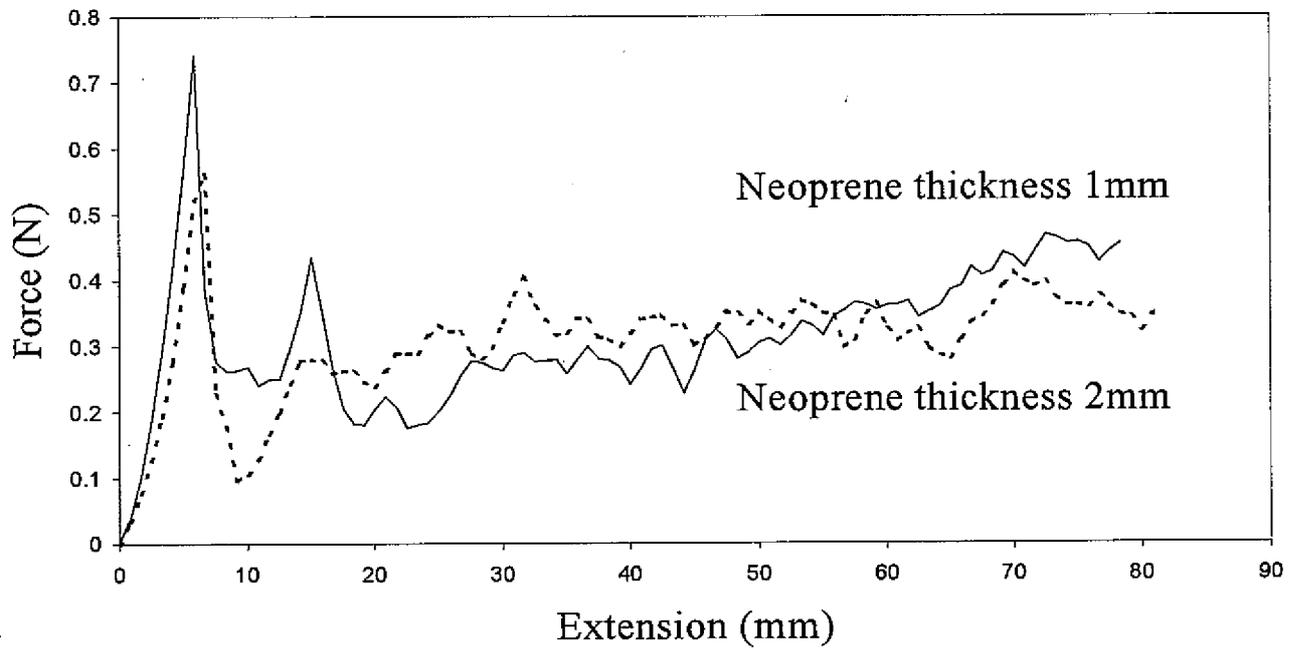
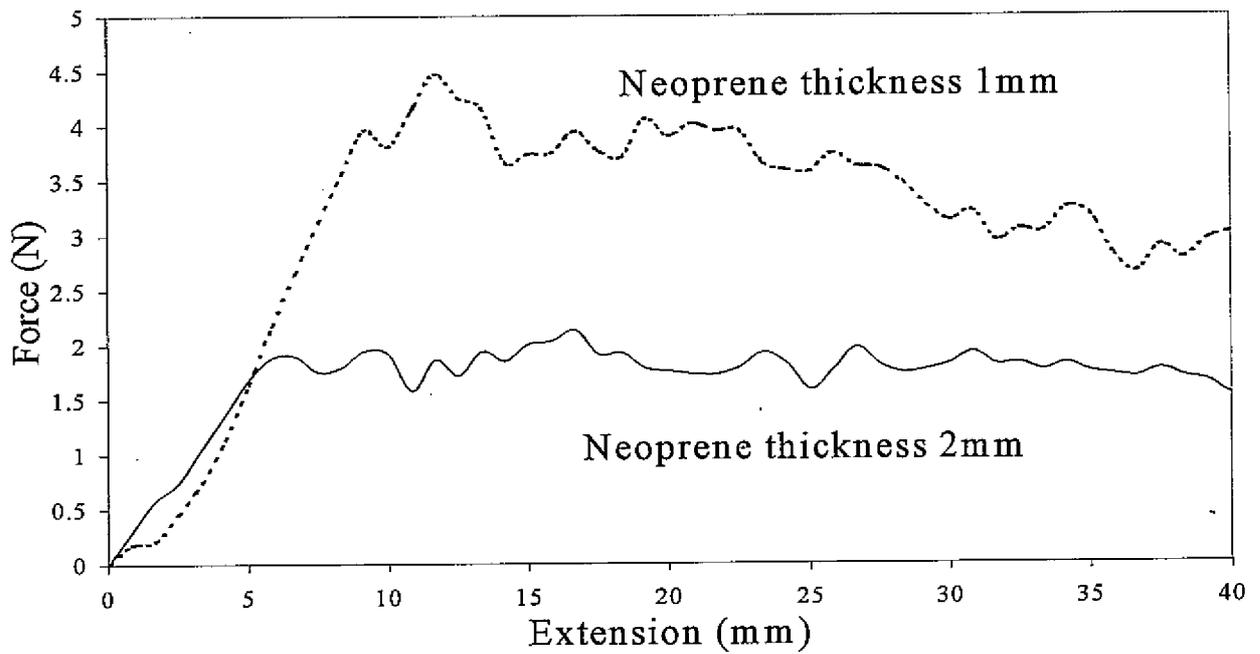
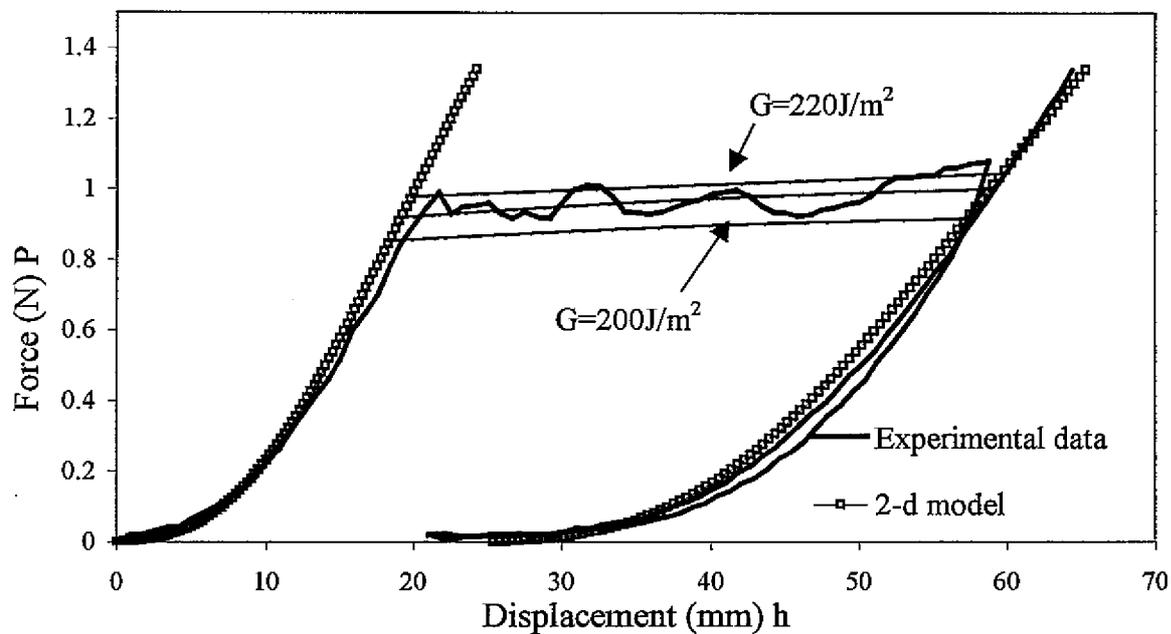


Figure 2b: Melinex peel test from neoprene rubber of different thickness, at a speed of 50mm/min, and nominal peel angle of 90°



An example of an experimental peel test is shown in Figure 3, where Melinex backing tape is peeled from 0.76mm thick latex. Also indicated on Figure 3 is the corresponding load and unload curves predicted by the 2-d model, for these specific test conditions, after a fracture length of 20mm. The experimental P-h trace shows that propagation occurs at an approximately constant  $G$  of about  $220 \pm 10 \text{ J/m}^2$ .

Figure 3: Experimental load-unload curves after 25mm fracture length for peeling Melinex from latex 0.76mm thick, with model prediction load-unload curves, for  $\nu=0.125$ .



The adhesive fracture energy (fracture toughness) of tapes during unpeeling is customarily calculated from the relation " $G = \frac{P}{b}(1 - \cos\theta)$ ". We show that while this expression is correct for peeling from rigid substrates, it gives misleading results when peeling from flexible substrates. A two-dimensional analysis is presented for peeling from non-linear elastic substrates that give consistent fracture energies from experimental data.

## Using Molecular Modelling as a Practical Aid to Adhesive Design

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### Abstract

The practical use of a combination of molecular modelling and structure-property relations is outlined for adhesives, with particular reference to pressure sensitive adhesives. The subject is taken here in two parts: the role of intermolecular forces in adhesive interfaces and compatibility of resin components, and the role of macromolecular structure and composition in thermal and mechanical viscoelastic properties.

### Introduction

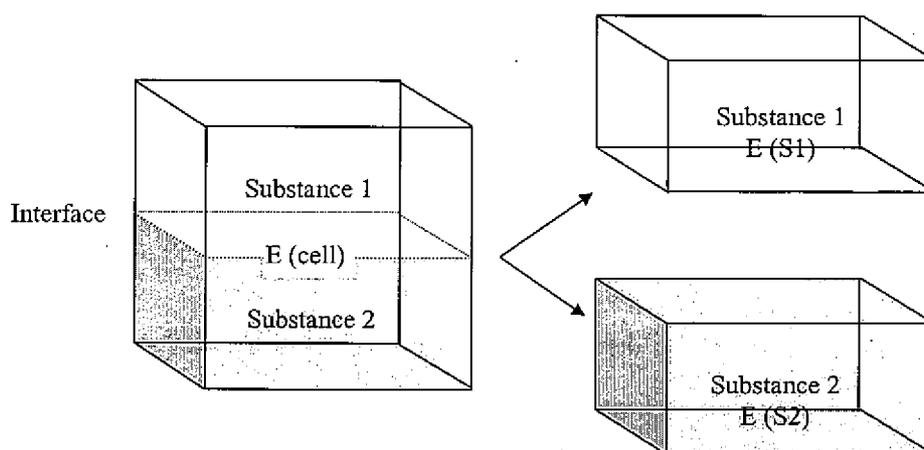
Although molecular modelling is now used extensively in the polymer industry, its use in applications such as adhesion is still relatively limited. Perceptions such as high cost (computer hardware and software plus manpower) and limited relevance of molecular-level studies to bulk engineering properties of materials are common. However, molecular modelling can be done on a desktop PC with a modest outlay on software [1]. Also, the term 'modelling' can be applied to everything from *ab initio* quantum mechanics simulations to totally empirical jottings on the back of a virtual envelope. Methods such as group contributions [2] and connectivity indices [3] are empirical structure-property tools for polymers that are easy to use. The method Group Interaction Modelling [4] of the author is discussed below, and links molecular-level parameters derived from atomistic simulations or empirical tables to bulk thermal and viscoelastic properties needed for understanding polymeric adhesives.

Although commercial molecular modelling software for polymers [5] can give excellent predictions of a limited range of physical properties, the direct translation of molecular modelling to application properties is still weak. This paper shows how a pragmatic combination of molecular modelling, polymer science, and practical experience can be used effectively as a working tool to facilitate adhesive design for specific applications and environmental conditions.

### Interfaces and Compatibility [6]

Most PSA applications require a good physisorption bond to be established between adhesive and substrate, with no permanent chemical bonding. Work of adhesion can be calculated using the scheme shown in Figure 1, where characteristic molecular cells of the two materials at an interface are created. The cells are fused by a process of molecular dynamics and energy minimisation. Work of adhesion is calculated simply the difference in energy per interface area between the fused and separated cells.

As a general comment on molecular modelling, the most important point is to ensure that the potential function used to calculate intermolecular energy is suitable for the materials of interest. Our experience shows that the Dreiding forcefield using Gasteiger charge assignment generally works well for modelling adhesion, and includes all the dispersion, polar, and hydrogen bonding components of energy.



*Figure 1. Calculating work of adhesion.*

Often, poor work of adhesion is due to mismatch in the polar components of energy. It is particularly important to assign the correct charges on inorganic substrate surfaces, which usually consist of metal oxides. Fixing atom position and assigning half the formal charge to the atoms at the substrate surface can give good estimates of the very high polar component of energy. This can be tested using interfaces with small molecule test fluids such as water, methyl iodide, and decane, just as in experimental measurements of surface free energy.

It is also possible to estimate the water sensitivity of a joint by applying thermodynamic rules with calculated values of surface free energy, with and without water. Adhesion promoters such as silanes simply act as bifunctional surfactants to match the energy between adhesive and substrate. Calculations have even been made using quantum dynamics to demonstrate that chemical bonds can be formed at a silanol-metal oxide interface.

Since PSA formulation often involves blending of two different polymer components, the compatibility of the components can be estimated reliably using solubility parameters, which are the square root of the cohesive energy density of each component of the intermolecular binding energy. These can be calculated simply using molecular modelling from the difference in energy between an isolated polymer chain and the same chain in an energy minimised solid cell. Alternatively, empirical group contribution tables or connectivity indices methods also give good results, since entropy of mixing plays little role in most polymer miscibility problems. More specific modelling tools have been developed to predict miscibility and include effects such as entropy and molecular weight effects [6]

### **Thermal and Mechanical Properties**

A key property for PSAs is their tack, which is due to a complex combination of viscoelastic properties. These viscoelastic properties must pertain at the application temperature, which is controlled by the glass transition temperature distribution of the polymer. The viscoelastic properties themselves are controlled by the relaxation time distribution and elastic modulus of the polymer. These are dictated both by temperature relative to the glass transition,  $T_g$ , and the molecular weight distribution. The molecular origin of these properties can be modelled using a combination of molecular simulations and Group Interaction Modelling. Here, the polymer is characterised by a small set of molecular level structural parameters: van der Waal's volume,  $V_w$ , cohesive energy,  $E_{coll}$ , chain stiffness,  $\theta$ , and thermodynamic degrees of freedom,  $N$ . These parameters are then used in a series of straightforward structure-property relations to predict bulk physical properties.

For example, the average value of  $T_g$  at a reference frequency of 1 rad/s is calculated using

$$T_g = 0.224 \cdot \theta + 0.0513 \cdot \frac{E_{coh}}{N}$$

The distribution and width of the glass transition zone is calculated using a power law probability distribution of pairwise interaction energies in the component concentrations.

Viscoelastic properties of PSAs are often complicated by being multi-component polymers, where crosslinked and thermoplastic components must be considered separately.

The key viscoelastic parameter is the macromolecular relaxation time,  $\tau_p$ , which is controlled by  $T_g$  and the molecular weights of the resin components. The scaling rules for  $\tau_p$  of the thermoplastic component follow the general proportionality

$$\tau_p \propto M_w^3 \cdot \exp\left(\frac{1590}{T - T_g + 50}\right)$$

For example, polyisobutylene has  $\tau_p \approx 0.1$  s with  $M_w = 55,000$ .

The elastic modulus of a high molecular weight polymer above its  $T_g$  is calculated from the energy remaining after the energy dissipation processes through the glass transition zone. The elastic modulus as a function of time around  $\tau_p$  is particularly important, since the resin 'flows' at times greater than  $\tau_p$ , but become more elastic at shorter times or at deformation rates greater than  $1/\tau_p$ . The viscosity of a polymer can be estimated from the product of elastic modulus and  $\tau_p$ . More detailed consideration of resins with high and low molecular weight components allows the flow of the PSA to produce the initial contact bond and the elastic reluctance of the resin to peel away from a substrate to be modelled.

The strength of a PSA is usually dictated by the long-time elastic modulus of the resin, which is usually the crosslinked rubber component or the high molecular weight fraction. The long-time modulus can be shown to have an approximately squared relation with the crosslinked fraction concentration, such that modulus would reduce approximately by a factor 4 at times greater than  $\tau_p$  if the fraction of crosslinked rubber is 0.5. Energetic considerations can be used to model cavitation processes in a polymer, which give relations similar to the Griffith crack initiation criteria. Such models could be used to quantify the fibrillation effects in PSAs during debonding in terms of the fibril dimensions, the elastic modulus of the resin, and the applied stress.

In summary, molecular modelling of PSA systems can provide valuable quantitative insights into their formulation and application properties that makes it valuable for resin design.

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## Polymer Resins for Waterborne Pressure-Sensitive Adhesives: Challenges for Industrial R & D.

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Tapes and labels are the two strategic groupings for pressure-sensitive adhesives (PSA) market (see Tab. 1). Tapes are mostly known in packaging and consumer/office applications. Labels are mainly recognised as decorative, promotion and information media. However, both types of products are also well present in other fields of application. Indeed, tapes are now applied in automotive and aerospace industries (to replace nails, for example), in electronics, in insulating and sound-deadening installations as well as in medical dressing. Specialty labels are used for security functions, in mailing, in data and goods tracking as well as in production control systems. Large labels for outdoor sign & marking applications, such as road marking, automotive and plane decoration and window advertising, are grouped in a special market segment: Graphic Arts. Finally, PSA are also used as masking, decorative, protective or overlaminating films, in envelopes...

**Tab. 1: General Overview of the European PSA Market ( $10^6$  m<sup>2</sup>)**

	2000		2003		Growth (%/Year)
	10 <sup>6</sup> m <sup>2</sup>	(%)	10 <sup>6</sup> m <sup>2</sup>	(%)	
<b>Tapes</b>	5850	54.5	6420	54	3
<b>Labels</b>	4020	37.5	4575	38	6.6
<b>Graphic Arts</b>	329	3	372	3	5
<b>Others (*)</b>	537	5	598	5	3
<b>Total</b>	10736	100	11965	100	4.5

(\*) masking and/or decorative films, protective or overlaminating films, envelopes...

In this growing market, the main driving forces are environmental considerations, cost-cutting concerns and improvement of PSA performances toward higher added-value products. These represent crucial challenges for polymer resin manufacturers. The development of the PSA market and ability to capture new areas of application depend on their capacity to tackle them.

As it is true for the whole adhesives industry, polymer resin manufacturers are strongly affected by changing environmental regulations. These companies are constantly tweaking their recipes to comply with new and existing environmental issues. Perhaps the most common trend, resulting from tighter restrictions by E.U. and U.S. agencies on the amount of volatile organic compounds (VOCs), is the move away from solvent-borne formulations to more "environmentally friendly" technologies such as waterborne PSA, and, to a smaller extent, UV-PSA. Nonetheless, switching to water-based systems is not always possible. Indeed, drying waterborne adhesives generally requires different coating conditions: i.e. higher temperatures and longer times (slower coating speed or longer ovens). Also several performance characteristics of solventborne PSA can not yet be reached with water-based systems: i.e., water and whitening resistance, high tack and adhesion. Over time, however, waterborne adhesives with equivalent performances and coatability will be available on the market.

Environmental and health aspects also impose in the choice of raw materials. For example, surfactants of the alkyl phenol ethoxylate type are now being banned from PSA recipes. Furthermore, for some label applications, there is an increasing demand of end users for systems containing chemicals, which are listed by health agencies, like FDA in the United States and BGVV in Germany.

Limitations in raw materials also come from cost-cutting concerns. Indeed, companies are looking towards simplifying the supply chain management of manufacturing. These considerations are, among others, strong driving forces to develop and optimise the product performances by controlling the morphology of latex particles as well as molecular mass and architecture of the polymer chains.

Another challenge is the sophistication and the diversification of product profiles of PSA required by the market. An example of higher demand in PSA performances are the requirements for higher adhesion on low-polarity surfaces and for improved water resistance. These follow the increase in market share of clear labels and the trend to replace PVC labels (mainly for environmental reasons) and paper labels by polyolefine labels. On the other hand, the diversification of application fields for PSA requires that new factors are taken into account in the design of PSA, like, e.g., the heat resistance in electronics, the sterilisation resistance and the water vapour permeability in medical applications.

Ensuring the success of a product also requires addressing the processing of the material. Here, one must take into consideration some economical aspects, such as high solids contents and high coating speeds (actually, up to 600 m/min for waterborne PSAs for paper labels, while there is a tendency to go to 1000 m/min) and high quality standards, which impose quick drying, clarity and smoothness of the film.

For R&D departments, these challenges motivate further fundamental studies of 'core' aspects of polymer latex technology, like, for example, latex particle morphology, latex film formation process and latex film microstructure, molar mass distribution, surfactant selection, etc. Their importance in determining PSA properties (adhesion, tack, shear) must be investigated and their action optimised. Beside this closer look to latex 'core' technologies, industrial R&D labs must open up to new ('emerging') technologies, which will allow one to overcome limitations of the presently used emulsion polymerisation techniques. To this end, miniemulsion and controlled/living radical polymerisation are promising, while the development of high throughput screening tools used in a combinatorial approach will accelerate new materials discovery. There, it is crucial for final success to take into account up-scaling aspects, from the beginning of the study.

## Pressure Sensitive Adhesives and Process Change

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**SCI Meeting - Adhesion Group**

**Tony Jones  
Harlow Chemical Company Ltd  
25<sup>th</sup> April 2001**

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- **Water-based pressure sensitive adhesives**
- **Components of Emulsion Polymerisation**
  - **Monomers, Emulsifiers, Initiators and Modifiers**
- **Manufacture**
  - **Typical industrial process**
- **Associated Common Problems**
  - **Film quality and Monomer Reactivity**

## Emulsion Polymerisation Components

Emulsion polymerisation is a free radical process to produce stable polymer dispersions.

For use as pressure sensitive adhesives, the resulting dried films must be inherently tacky.

### **Monomer**

Produces polymer particles e.g. Butyl acrylate, Styrene

### **Emulsifier**

Prevents coagulation e.g. alkyl sulphate

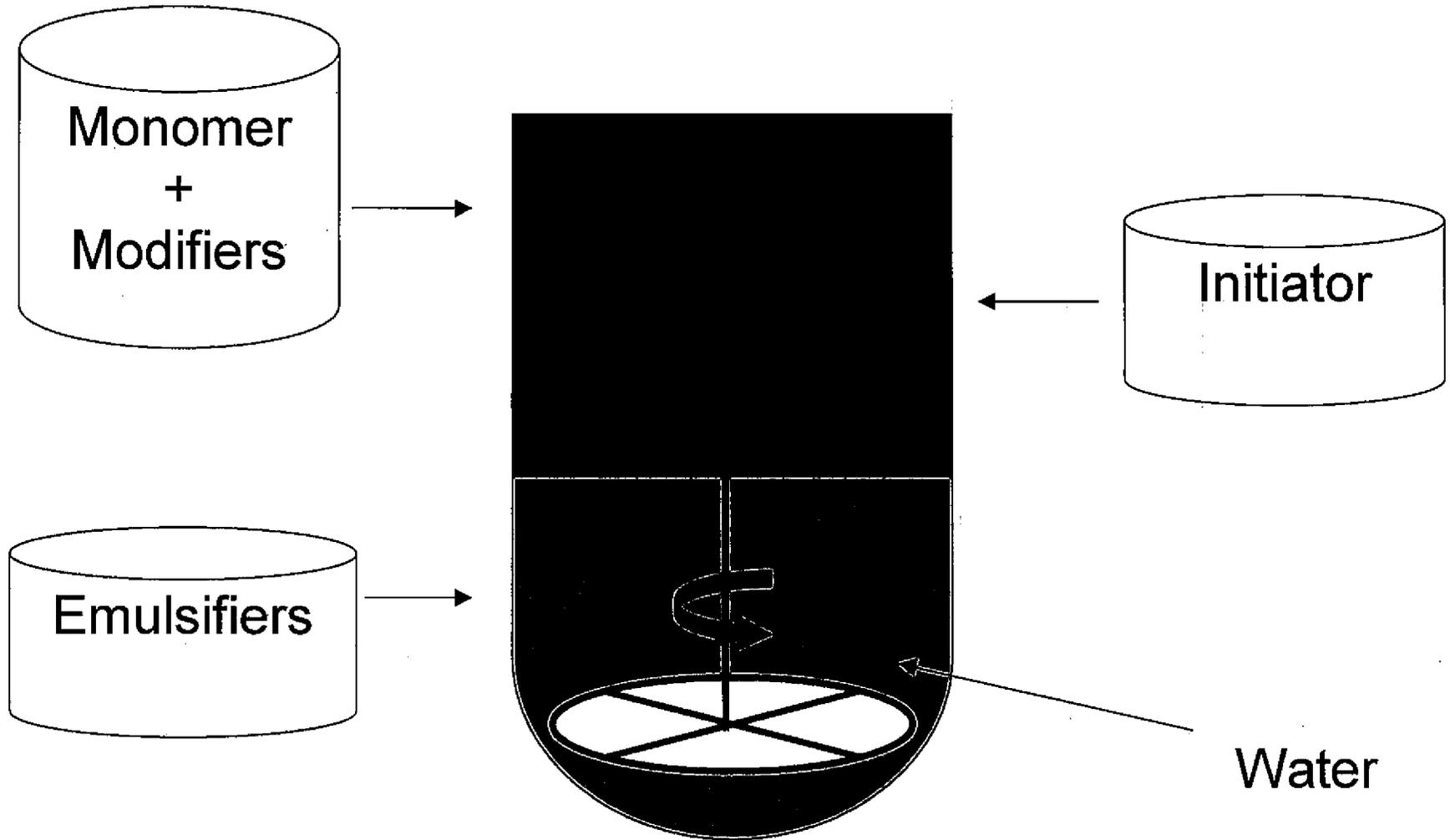
### **Initiator**

Starts polymerisation e.g. sodium persulphate

### **Modifiers**

Changes polymer properties e.g. Chain transfer agents

Manufacturing Process (typical)



# Water-Borne Pressure-Sensitive Adhesives

## *Correlation of Properties with Particle Structure*

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### **Abstract**

Environmental pressures, together with increasingly stringent legislation concerning solvent emissions, have forced the coatings industry to seek the replacement of solvent-borne adhesives by all-solids coatings and/or water-borne coatings. These pressures have been particularly strong in Europe and North America [e.g., 1] and, as a consequence, interest in water-borne coatings is growing substantially. Because conventional solvent-borne coatings typically are based on water-insoluble polymers, aqueous dispersions are required in order to use them for water-borne coatings. Emulsion polymerisation is a particularly convenient means of preparing such hydrophobic polymers in a form suitable for formulation into water-borne coatings, the principal coating types prepared in this way being paints and adhesives [2]. There has, therefore, been growing commercial interest in coating materials prepared by emulsion polymerisation.

The effect of this growth in demand for water-borne systems on the adhesives market is most evident for pressure-sensitive adhesives and contact adhesives, since these types of adhesive have traditionally been dominated by solvent-borne systems. In contrast, adhesives for use with wood and board have for many years been based on poly(vinyl acetate) latexes and so have not been so markedly affected.

This paper will present an overview of the preparation and properties of water-borne adhesives based on n-butyl acrylate [3]. Correlations between latex particle composition/morphology, polymer properties and adhesive performance will be outlined with particular emphasis on the effects of particle morphology and the way in which acrylic acid is used in the emulsion polymerisations.

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