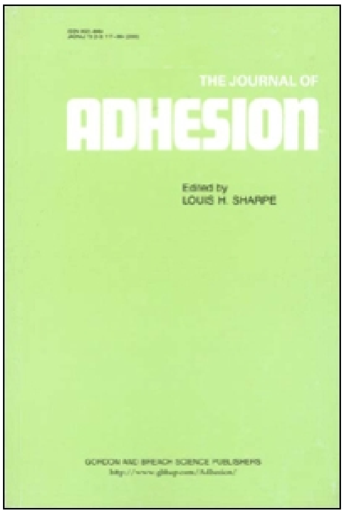

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Evaluation of the Constrained Blister Test for Measuring Adhesion

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The constrained blister test (CBT) was evaluated as a method for measuring adhesion using a model system, electrical tape bonded to polystyrene. Pressure is applied through a circular inlet hole in the substrate, causing the adhesive to "blister" up and peel radially away from the substrate. A glass constraint, placed some distance above the adhesive, limits deformation of the adhesive in the vertical direction and promotes radial peel. By operating at low spacer height (the distance of the constraint above the adhesive) and very low growth rates, the energy spent for deformation of the adhesive and viscoelastic dissipation is minimized. Blister radial growth was linear with time, and growth rate increased linearly with the second power of the energy input. An intrinsic, rate-independent adhesion energy was obtained by extrapolation to zero crack growth rate. The CBT was compared with two peel tests. The dependence of the growth rate on energy input was different, but the extrapolation to zero growth rate gave the same value of the intrinsic adhesion energy.

Keywords: Constrained blister test; adhesion; intrinsic adhesion; polymer interface; peel test; detachment energy

INTRODUCTION

The adhesion energy, G, measured in dynamic tests that require peeling of the adhesive consists of two components [1,2]:

$$G = G_c + \psi$$

*Corresponding author.

where G_c is the intrinsic energy of adhesion to debond the two surfaces, which is independent of test geometry and debonding rate, and ψ, which includes all other sources of energy consumption, *i.e.*, the energy spent deforming the adhesive and/or substrate, and the energy viscoelastically dissipated in the damage zone ahead of the interfacial crack. The ψ term depends on test geometry, temperature, and the crack growth rate, and can often be orders of magnitude greater than G_c. For the same adhesive-substrate system, different adhesion tests may yield very different values of the adhesion energy, G, because of different ψ contributions. Therefore, adhesion tests that measure G_c are very desirable.

The constrained blister test (CBT), shown schematically in Figure 1a, may operate under conditions where the ψ component is negligible [3–6]. The constraining plate minimizes deformation of the adhesive, and the pressure can be adjusted so that the test runs at very slow rates to minimize the viscoelastic energy dissipation in the damage zone. The constrained blister test has several advantages over the unconstrained blister geometry, which has been studied by several authors [7–10]. The constraining plate limits deformation of the adhesive, so adhesion can be measured in systems where the adhesive strength is on the same order of magnitude as the cohesive strength. Also, in the constrained blister test the energy input to the blister can be nearly constant with time, and the steady state blister growth rate for a given input energy can be measured. In contrast, only the critical pressure to cause crack propagation is measured in the unconstrained geometry. The CBT also has several advantages over peel tests. The standard *T*-peel test may cause large deformation of the arms, which usually ensures a large ψ component, and essentially prohibits testing of brittle materials. The "blistering" mode of failure mimics real life failures of paints, coatings, laminates, and adhesives. Furthermore, the test geometry allows easy examination of environmental effects by pressurizing the system with different fluids.

Despite an increasing interest in the CBT, only a few experimental studies of adhesion by this method are presented in the literature [3–5]. No reliable procedure for obtaining an intrinsic adhesion value from blister growth has been established. The simplified model used for this purpose in previous studies [3, 4] predicted an exponential increase of the radial blister growth rate with time, which is inconsistent with the constant growth rate observed in experiments

[4, 5]. Also, the effect of pressure and spacer height on the stability of blister growth has not been investigated. The goal of this work was to examine the validity of the constrained blister test in a wide range of experimental parameters in order to determine conditions whereby an intrinsic adhesion energy, G_c, could be obtained. The G_c value was compared with that determined in peel tests. A model system of an electrical tape bonded to polystyrene (PS) was used in this study.

THE CONSTRAINED BLISTER TEST

In the constrained blister test, Figure 1, the pressure is applied through a circular inlet hole in the substrate, causing the adhesive to "blister" up and peel radially away from the substrate. A glass constraint,

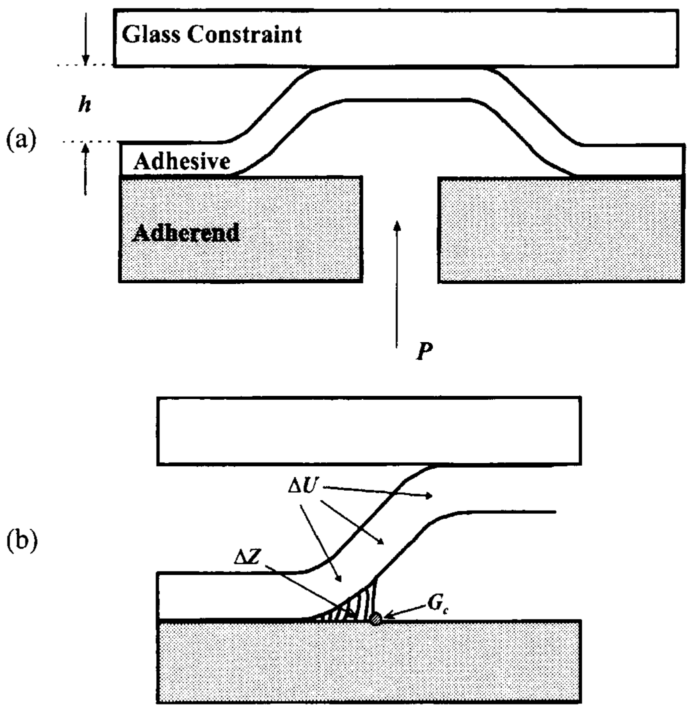

FIGURE 1 Schematics of the constrained blister test: (a) side view of the constrained blister test; and (b) contributions of G_c, the intrinsic adhesion energy; ΔU, the energy spent deforming the adhesive; and ΔZ, the viscoelastic energy dissipated in the damage zone.

placed some distance above the adhesive, limits deformation of the adhesive in the vertical direction and promotes radial peel. The distance between the adhesive and the glass constraint is determined by the height of the spacer, h. The pressurizing medium is N_2 gas, and pressure is controlled with an accuracy of ± 0.25 psi (1.72 KPa). Blister growth as a function of time is monitored by video cameras from the top and side. Typical images of a growing blister are shown in Figure 2. Top view images are stored on computer, and an image analysis program is used to measure the detached blister area and the area contacting the constraint. If the blister is nearly round, the area can be used to compute an average detached radius, r_d, and an average contact radius, r_c. The detached radius, r_d, is the distance from the center of the inlet hole to the point of detachment from the substrate. The constrained radius, r_c, is the distance from the center of the inlet hole, juxtaposed to the glass constraint, to the point of contact of the tape with the constraint, Figure 2. The difference between r_d and r_c is the suspended distance, l. The detachment angle, θ, defined as the maximum angle of the tape between the two bending regions, is measured from the blister profile with an accuracy of $\pm 3°$.

ENERGY INPUT TO THE CONSTRAINED BLISTER

The energy balance in the CBT is [3–6]:

$$\Delta W = G\Delta A = G_c\Delta A + \Delta U + \Delta Z$$

where ΔW is the work to debond an area, ΔA; ΔU is the change in the potential energy of elastic deformation; and ΔZ is the viscoelastic energy dissipated in the damage zone per area debonded, ΔA. In the CBT, G is obtained experimentally, and test conditions are sought that minimize ΔU and ΔZ in order to obtain G_c.

The input energy per unit surface area created, $G = \Delta W/\Delta A$, is equal to $P\Delta V/\Delta A$, where ΔV is the change in blister volume. The quantity ΔV is not easily measured, but can be calculated by assuming a blister shape. A blister shape factor, $q \equiv (1/h) \cdot (dV/dA)$, is usually introduced so that $G = qPh$ [5, 6]. By assuming the blister shape to be that of a conical section of height, h, the blister shape factor, q, is

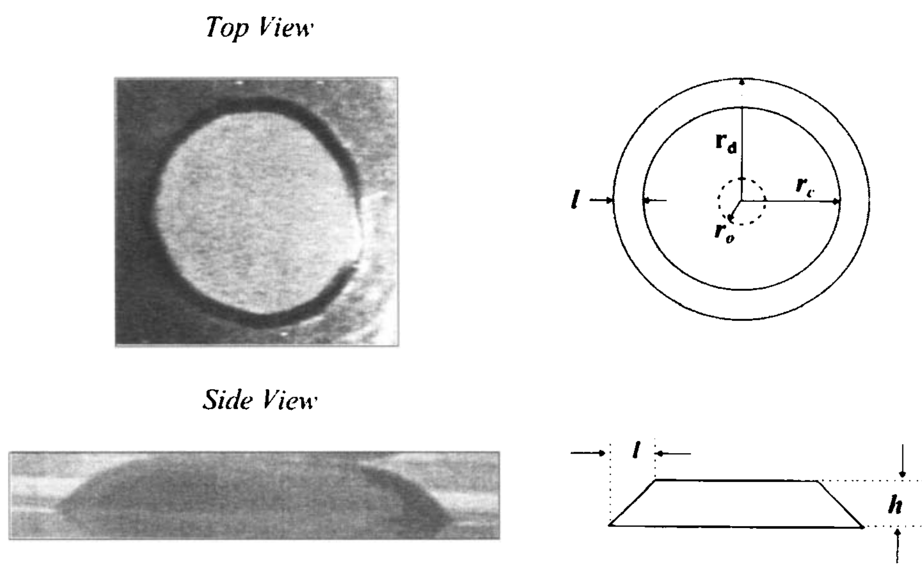

FIGURE 2 Photographs showing top and side views of blister growth, and accompanying schematics illustrating parameters measured in the test: r_d is the detachment radius, r_c is the constrained radius, l is $r_d - r_c$ or the suspended distance, r_0 is inlet hole radius, and h is the spacer height.

calculated to be:

$$q = \frac{1}{h} \cdot \frac{dV}{dA} = \left(1 - \frac{l}{2r_d}\right) + \left(\frac{l}{3r_d} - \frac{1}{2}\right) \cdot \frac{dl}{dr_d} \tag{1}$$

For a detachment angle of 90° (cylindrical blister) $l = 0$, $dV/dA = h$, and $q = 1$. For a detachment angles less then 90°, q is less than one but asymptotically approaches 1 at large r_d. Experimentally, l changes little with r_d, and as a result the second term in Eq. (1) is two orders of magnitude less than the first term and can be considered negligible. A test where q is equal to or close to unity is desirable so that the normalized energy input $G = qPh$ is nearly constant throughout the test.

EXPERIMENTAL SYSTEM

The substrate was Styron 685D polystyrene (PS) from Dow Chemical Company. Pellets were placed in a 4.5 mm thick mold between two steel plates with a mirror finish. The mold was preheated in a press for 10 minutes at 175°C. A pressure of 300 psi (2.1 GPa) was applied at this temperature for an additional 10 minutes, and then the mold was cooled under pressure to room temperature at about 10°C/min. A 3 mm diameter inlet hole was machined into the PS, and a pressure sensitive adhesive tape, Super 33 + electrical tape 0.175 mm thick manufactured by 3 M, was applied to the substrate with finger pressure. The sample was held under a pressure of 5 psi (34 KPa) for two minutes, and left overnight before testing. Because of differences in adhesion between rolls of tape and aging effects, all constrained blister tests were performed with tape from a single, new roll. The temperature was 19.5 ± 0.3°C.

The deformation response of the adhesive was characterized with ASTM D1708 microtensile specimens cut from the electrical tape. Specimens were deformed in an Instron 1123 testing machine at different strain rates (Fig. 3a). Stress-strain behavior upon consecutive loading and unloading at the same strain rate is presented in Figure 3b. The material showed typical viscoelastic behavior with almost completely reversible deformation. The viscous contribution decreased

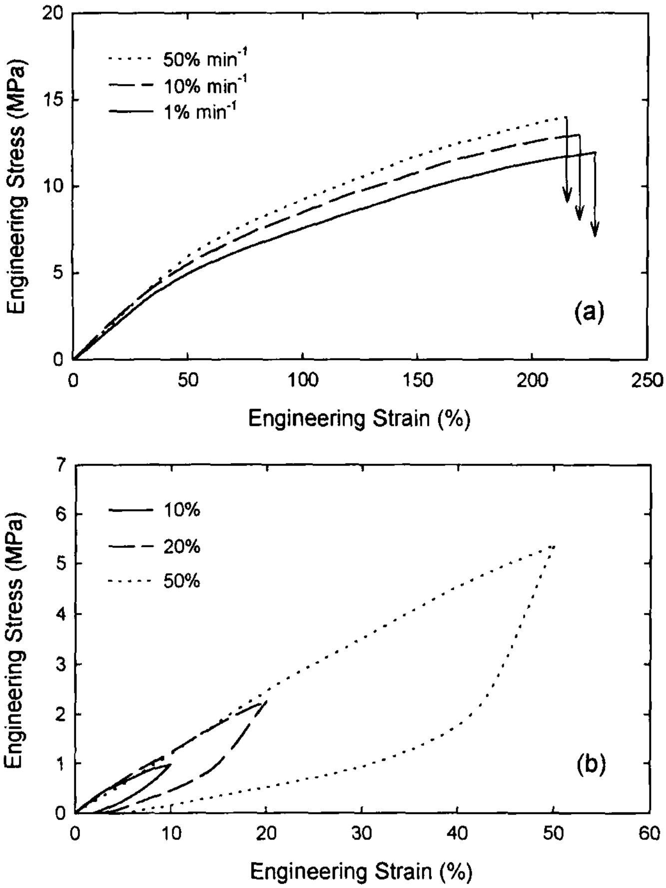

FIGURE 3 Stress-strain behavior of the adhesive tape in uniaxial tension: (a) deformation at different strain rates; (b) loading–unloading cycles to different strains at strain rate 10% min^{-1}.

with decreasing strain, and deformation in the range below 10% could be considered as elastomeric. Deformation in the CBT, estimated to be on the order of the ratio h/r_d, usually fell into this range.

Growth rates on the order of 0.1 cm/min were achieved using Ph values in the range of 30 to 80 J/m^2. For convenience, combinations of P and h that gave $Ph = 71$ J/m^2 were used in many of the tests to evaluate the CBT with the electrical tape/PS system. In Figure 4a, three trials are shown for one combination of pressure and spacer

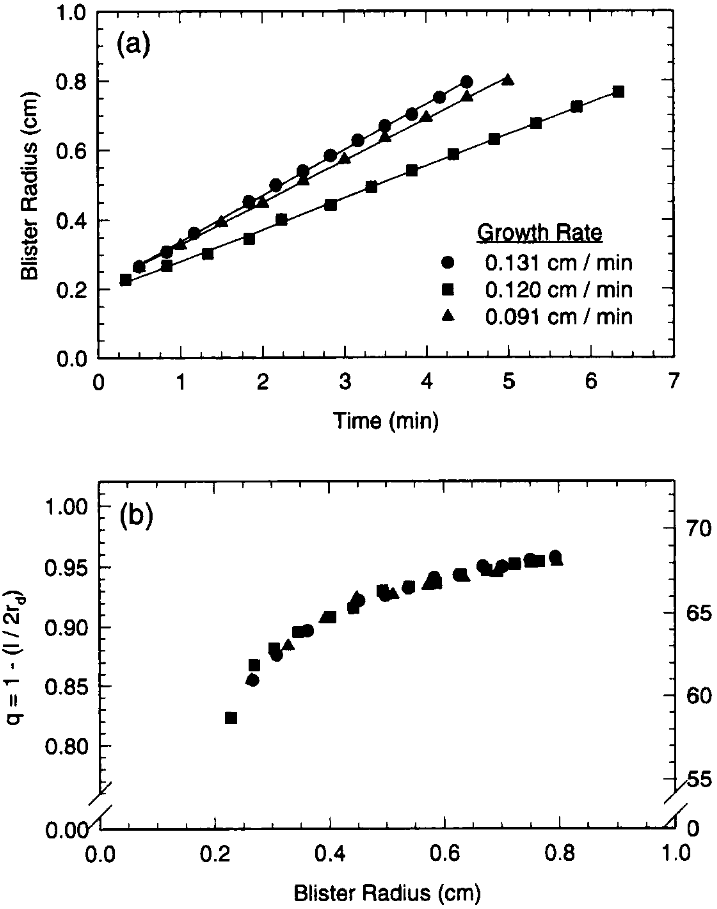

FIGURE 4 Three CBT trials at energy input (Ph) of 71 J/m^2 where P = 27 psi (188 KPa) and h = 0.38 mm: (a) blister growth; and (b) blister shape factor, q. The average growth rate was 0.11 cm/min and the standard deviation was 0.02.

height, 27 psi (185 KPa) and 0.38 mm. The data are plotted as time *vs.* the average detachment radius, r_d. The increase in r_d was linear with time, as reported previously [4, 5]. Because the radial growth rate was independent of blister size, this parameter was chosen to characterize blister growth. For this value of Ph the average radial growth rate was 0.11 cm/min, the standard deviation was 0.02.

The change in the blister shape factor, q, with blister growth is shown in Figure 4b for the three trials in Figure 4a. Initially q was about 0.7. It asymptotically approached one at large blister radius, and toward the end of the test, q was about 0.95. The change in q with blister growth was nearly the same in each trial.

EVALUATION OF BLISTER TEST PARAMETERS

Test conditions which minimized ΔU needed to be established. The energy spent deforming the tape, ΔU, has two components. A bending component acts in the two regions where the tape curves: in the detachment region and in the vicinity of the point of contact with the constraint. A stretching component acts between the bending regions where the tape is stretched. At low enough spacer height the stretching component becomes negligible and, for a soft adhesive, the bending component is nearly zero [11]. If the spacer height is too low, however, the constraint may impinge on formation and growth of the damage zone. The spacer height should be low enough that stretching deformation of the tape is minimal, yet high enough to allow stable blister growth and shape.

The effect of spacer height was characterized by holding Ph constant and varying h (and P). The effect of changing spacer height on blister growth for $Ph = 71\ \mathrm{J/m^2}$ is shown in Figures 5a and 5b. For the 0.26, 0.38 and 0.52 mm spacer heights, Figure 5a, the time for the tape to contact the glass constraint was less than 1 second after pressure was applied, and radial blister growth was linear over the entire test. The growth rate appeared to decrease slightly with spacer height in this spacer height range. Figure 5b shows blister growth for the 0.78 and 1.04 mm spacer heights. With these larger spacer heights, the tape did not contact the constraint immediately. The time for the tape to contact the constraint was 30 seconds for the 0.78 mm spacer height and 2.5 minutes for the 1.04 mm spacer height. After contact, the radial growth rate was constant. In another series of experiments with $Ph = 36\ \mathrm{J/m^2}$, the contact time of the tape with the constraint was again less than a few seconds when the spacer height was 0.52 mm and below, but then increased from about 25 minutes for the 0.78 mm spacer height to 150 minutes for the 1.04 mm spacer height. The

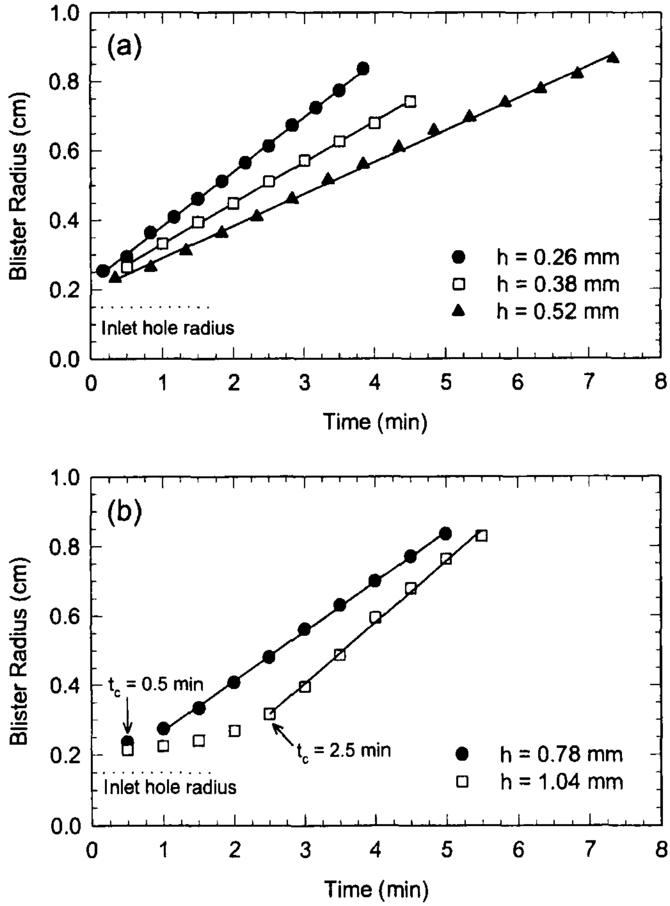

FIGURE 5 Effect of spacer height on blister growth at constant $Ph = 71\ \mathrm{J/m^2}$: (a) 0.26, 0.38, and 0.52 mm spacer heights; and (b) 0.78, and 1.04 mm spacer heights. The time at which the tape contacts the constraint is designated t_c.

presence of an induction time might have indicated that at these heights the tape experienced significant stretching that could have affected the blister growth rate.

The blister shape factor, q, was calculated from r_d and l. In Figure 6 the blister shape factor and the energy input, $G = qPh$, for the experiments in Figures 5a and 5b are plotted as a function of blister radius. Because $q = 1 - (l/2r_d)$, the blister shape parameters that determine q are the detached blister radius, r_d, and the suspended

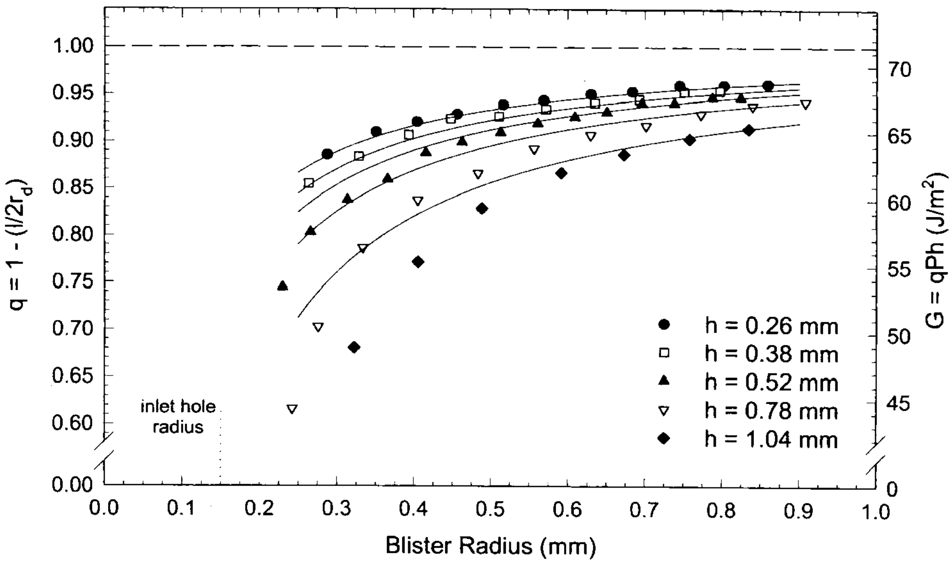

FIGURE 6 Effect of spacer height on blister shape factor, q, from measured values of the suspended distance, l (symbols); and calculated from l measured at the end of the test (lines).

distance, l. Increasing spacer height decreased q by increasing l. However, l decreased as r_d increased, and by the end of the test l was much less than r_d. As a result, q was between 0.92 and 0.96 for all spacer heights. The solid curves in Figure 6 correspond to the q values calculated using the l measured at the end of the test. The deviation from the measured q shows the effect of the changing l on q. For low spacer heights, l was very small and almost constant so the solid curve matched the measured points over the entire test. With increasing spacer height, l decreased more with increasing r_d, and the deviation from the solid curve became noticeable. Because q was between 0.92 and 0.96 at the end of each test, q was omitted from subsequent data analysis and the energy input was reported as $G = Ph$.

The effect of spacer height on detachment angle is shown in Figure 7. The angle increased from about 30° at the beginning of the test to 50° at the end of the test. The increasing detachment angle correlated with the decreasing l. For a given blister size, the angle increased with increasing spacer height in the 0.38, 0.52 and 0.78 mm spacers. The detachment angles were similar for the 0.78 mm and 1.04 mm spacers.

The radial growth rate as a function of spacer height for two energy inputs, $G = Ph = 71\,\mathrm{J/m^2}$ and $36\,\mathrm{J/m^2}$, is shown in Figures 8a and 9a. When the spacer height was greater than 0.26 mm, blisters grew in a nearly-circular shape with an essentially constant growth rate. Because the increase of h increased tape deformation, the constant growth rate indicated that the contribution of the elastic energy, ΔU, to the total energy consumption was negligible. The effect of stretching became perceptible only at the largest spacer height, 1.04 mm, which was 6 times the tape thickness. Blister growth rates for very low spacer heights, 0.085 to 0.26 mm, had a very large standard deviation. Visual inspection of the specimens after testing revealed that these blisters grew in a tunneling fashion, where adhesive material remained attached to the substrate behind the propagating crack front. Apparently, if the spacer height was less than the thickness of the tape, 0.175 mm, blister growth was unstable. The possibility of blister growth instability was considered in theory [12] by assuming the presence of residual stress in the adhesive. However, the shape of the unstable blister varied randomly with no correlation between the orientation of unstable growth blister and the adhesive tape rolling direction.

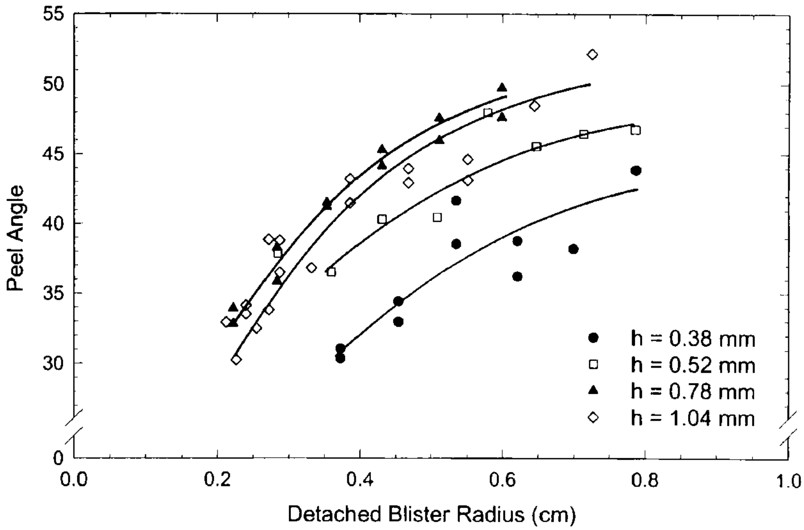

FIGURE 7 Effect of spacer height and blister size on peel angle.

Blister shape was characterized by the circularity, defined as the blister circumference squared divided by the blister area. The resulting quantity is dimensionless and is equal to 12.57 (4π) for a circle. Plots of circularity *vs.* spacer height are shown in Figures 8b and 9b. In the spacer height region of stable blister growth, 0.26 to 1.04 mm, the blister was nearly circular with low standard deviation. Below 0.26 mm the blister shape deviated markedly from circular, and the standard deviation increased. Based on the blister growth and shape data, the 0.26, 0.38, and 0.78 mm spacer heights were chosen for the experiments of varying *Ph*.

INTRINSIC ADHESION FROM CBT AND OTHER ADHESION TESTS

Operating at very low crack growth rates minimized ΔZ, the energy dissipated viscoelastically in the damage zone. The blister growth rate was varied by adjusting the energy input, *Ph*, to the blister. Figure 10 shows the results from experiments with three spacer heights and

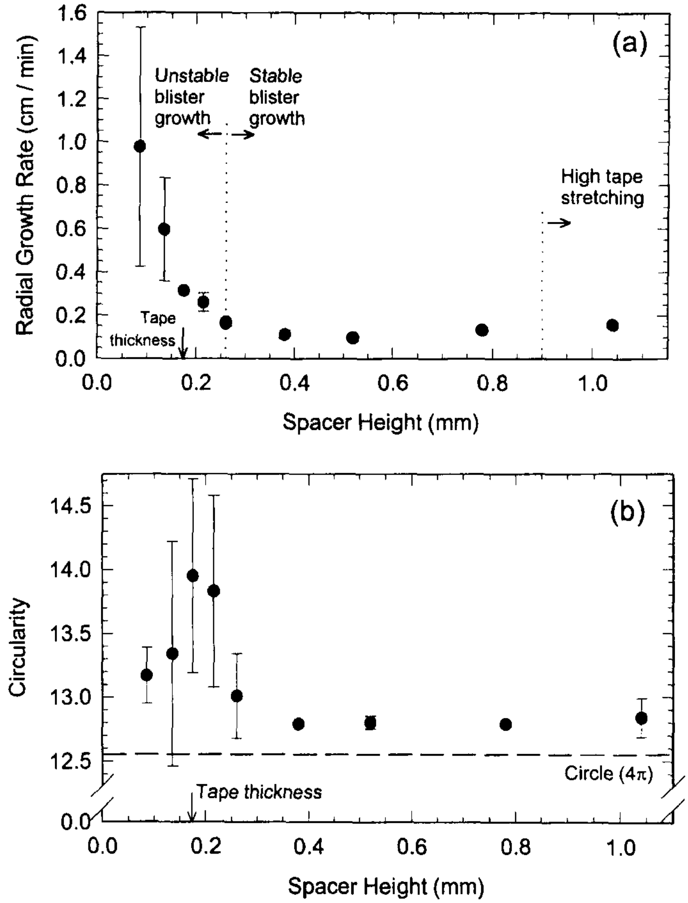

FIGURE 8 Effect of spacer height on blister growth for $Ph = 71\ \mathrm{J/m^2}$: (a) blister growth rate; and (b) circularity of blister shape, $(\mathrm{Circumference})^2/\ \mathrm{Area}$.

various pressures. The data are plotted as blister growth rate raised to the 1/2 power *vs.* Ph. The linear relationship extended over three orders of magnitude from a growth rate of nearly 0.35 cm/min for $Ph = 100\,\mathrm{J/m^2}$ to less than 0.0004 cm/min (4 μm/min) for $Ph = 12\ \mathrm{J/m^2}$. The solid line is a linear regression line through all the data.

The empirical relationship between energy input and crack growth rate made it possible to extrapolate to zero crack growth rate to determine G_c. The intrinsic adhesion obtained by extrapolation for

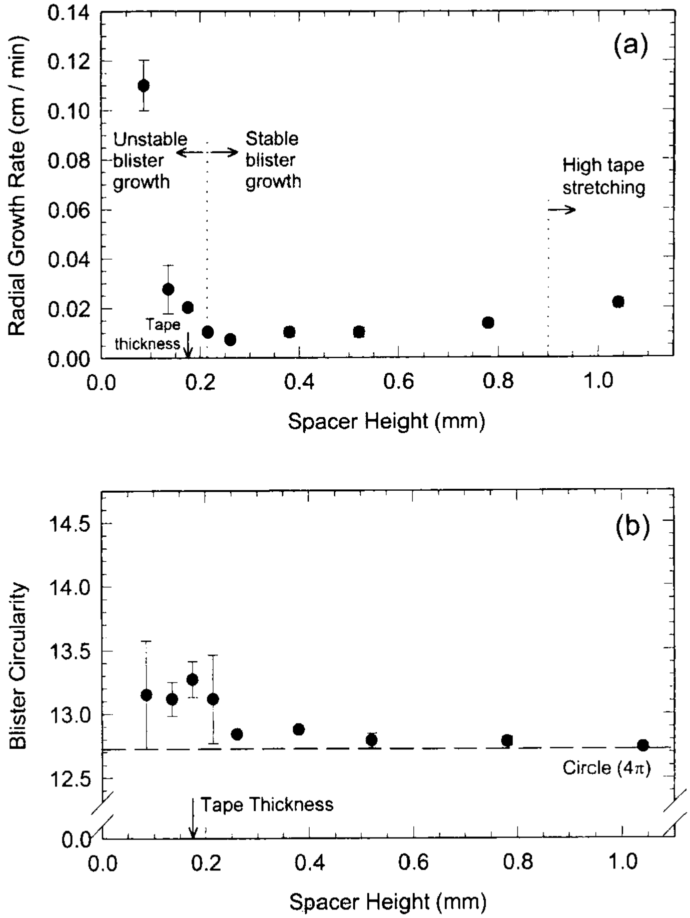

FIGURE 9 Effect of spacer height on blister growth for $Ph = 36$ J/m^2: (a) blister growth rate; and (b) circularity of blister shape, (Circumference)^2/ Area.

each spacer height was 12.8, 10.0 and 9.2 J/m^2 for the 0.26, 0.38, and 0.78 mm spacer heights, respectively. The average was 10.7 ± 1.9 J/m^2. The second-order fit described the data so well, in fact, that if the growth rate points for Ph less than 40 J/m^2 were removed, the fit through the remaining data still extrapolated to about 10 J/m^2.

Adhesion measurements made in peel tests for a range of crack growth rates were compared with the CBT data. Three peel tests were employed: 180° peel, 180° peel around a 200 g roller 12.7 mm in

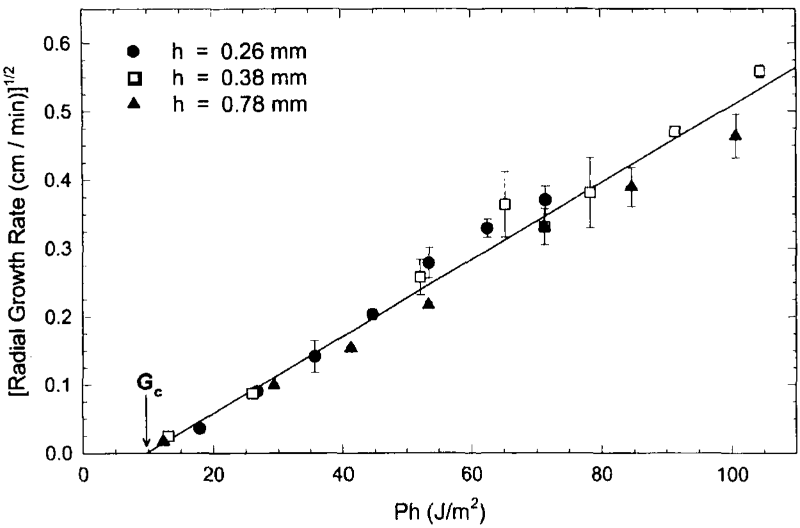

FIGURE 10 Effect of energy input, *Ph*, on blister growth rate showing the extrapolation to zero growth rate to obtain the intrinsic, rate-independent adhesion energy, G_c.

diameter, and the same 180° peel around the roller with the tape reinforced with Mylar™ to prevent stretching. Bending and stretching contributions differed in each test. In the 180° peel test, bending was high because the detachment angle was sharp, and stretching was low. In the peel around the weighted roller, bending was much lower as the detachment angle was decreased to about 60°, but the weight significantly increased stretching. Stretching was prevented when the tape was backed with Mylar sheet.

Figure 11 shows the effect of rate on peel adhesion energy compared with the CBT. At least 2 series of data were generated for each test geometry. In the peel tests the crack growth rate was fixed and the adhesion energy, G, was measured. In contrast, in the CBT the adhesion energy, Ph, was fixed and a crack growth rate was measured. Peel adhesion energy, G, was calculated from the relation $G = P(1-\cos\theta)/w$ where P is the load, θ is the angle from which the load is applied (180° in each test), and w is the specimen width [11]. As was the case in the CBT, there was a linear relation between peel crack growth rate and G^2. Neither tape bending nor stretching appeared to affect

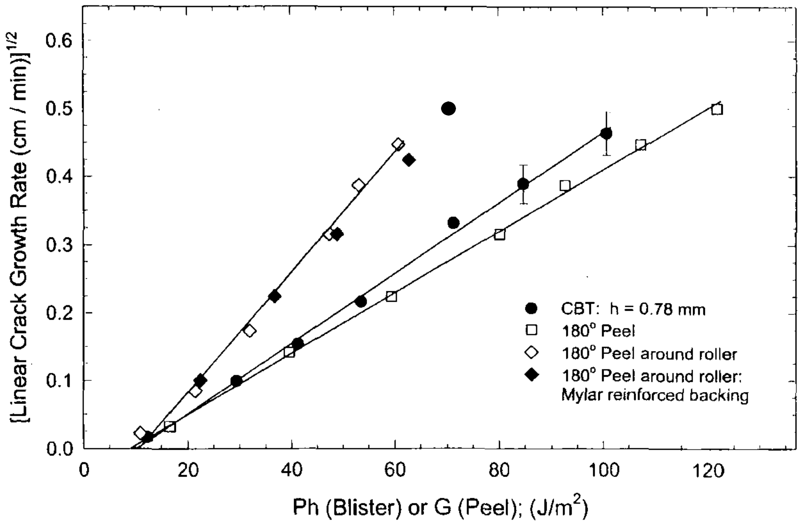

FIGURE 11 Comparison of the CBT with peel tests.

crack growth rate significantly. Preventing stretching by backing the tape with Mylar did not affect crack growth rate. Furthermore, bending did not appear to affect crack growth rate because the data from the 180° peel, where bending was high, correlated with the CBT, where bending was low.

Although the test geometry had some effect on the crack growth rate, extrapolation to zero growth rate yielded nearly the same values of G_c, the rate-independent intrinsic adhesion energy. The values were $8.8 \pm 0.2\,\mathrm{J/m^2}$ for the 180° peel test, $9.3 \pm 1.9\,\mathrm{J/m^2}$ in the 180° peel around the roller tests, and $10.7 \pm 1.9\,\mathrm{J/m^2}$ in the constrained blister test. The coincidence of G_c values obtained by different tests lends credibility to the extrapolation method used to obtain G_c.

The values of G_c obtained in both the CBT and peel tests are consistent with previous assessments of adhesion energies of commercial tapes bonded to different rigid substrates, which vary from 1.5 to $44\,\mathrm{J/m^2}$ [3, 4]. These values are significantly higher than the thermodynamically reversible work of adhesion, W_a, which is typically in the range of $0.05-0.1\,\mathrm{J/m^2}$ for immiscible polymer pairs [13]. The quantity W_a is related only to the surface energies of the two materials, and can be determined from contact angle measurements of liquid droplets on the surfaces. The difference between G_c and W_a for polymeric adherends is often ascribed to the modification of the interface due to interpenetration and entanglement of surface chains [14–18]. Pull-out of interfacially-anchored chains requires additional energy that can considerably exceed W_a.

There is another reason for G_c to be higher than W_a if the adhesion energy is measured by a method that requires peeling the adhesive. An interfacial crack is preceded by a damage zone of highly-deformed material which relieves the stress concentration at the crack tip. The damage zone forms even at vanishing crack growth rate, in which case it remains stationary within the time scale of interest. The work to form this stationary damage zone contributes to the threshold value of the adhesion energy obtained by extrapolation to zero crack growth rate. This contribution can be important for materials with good adhesion, including this electrical tape which was designed to have good tack to a variety of materials.

SUMMARY

In summary, the constrained blister test made it possible to minimize deformation of the adhesive so that the energy input was correlated with the interfacial crack propagation rate only. However, the constraint of the blister below a certain limit gave rise to instability of the blister growth. The range of experimental parameters was optimized for the testing of an electrical tape as an example of a typical elastomeric adhesive. A second-order relation between the blister growth rate and the energy input was empirically established, which permitted reliable extrapolation to zero growth rate in order to obtain an intrinsic adhesion energy, G_c. The values of G_c were consistent with values measured from peel tests.

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