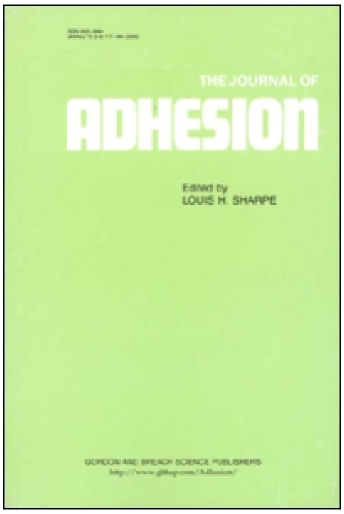
This article was downloaded by:
On: *22 January 2011*
Access details: *Access Details: Free Access*
Publisher *Taylor & Francis*
Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



The Journal of Adhesion

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

Micromechanics of Fracture in Structural Adhesive Bonds

D. L. Hunston[a]; A. J. Kinloch[b]; S. S. Wang[c]
[a] National Bureau of Standards, Polymers Division, Gaithersburg, MD, USA [b] Department of Mechanical Engineering, Imperial College, London, England [c] Department of Theoretical and Applied Mechanics, University of Illinois, Urbana, Illinois, USA

To cite this Article Hunston, D. L. , Kinloch, A. J. and Wang, S. S.(1989) 'Micromechanics of Fracture in Structural Adhesive Bonds', The Journal of Adhesion, 28: 2, 103 — 114

To link to this Article: DOI: 10.1080/00218468908030877

URL: http://dx.doi.org/10.1080/00218468908030877

PLEASE SCROLL DOWN FOR ARTICLE

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

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

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

J. Adhesion, 1989, Vol. 28, pp. 103–114
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United Kingdom

Micromechanics of Fracture in Structural Adhesive Bonds

D. L. HUNSTON

National Bureau of Standards, Polymers Division, Gaithersburg, MD 20899, USA

A. J. KINLOCH

Imperial College, Department of Mechanical Engineering, London SW7 2BX, England

S. S. WANG

University of Illinois, Department of Theoretical and Applied Mechanics, Urbana, Illinois 61801, USA

(Received March 11, 1988)

The high mode-I fracture surface energies, G_{IC}, of structural adhesives can be attributed to their ability to form large crack-tip deformation zones prior to failure. It has been suggested that this feature also controls the dependence of the adhesive bond G_{IC} on bond thickness. The proposed explanation asserted that the physical constraint of the adherents and the nature of the crack-tip stress field in an adhesive joint alter the size and shape of the deformation zone, and this in turn changes the fracture behaviour. To examine this hypothesis, motion pictures were taken of fracture specimens during loading, and the stress whitening that occurred at the crack tip was used to judge the relative dimensions of the deformation zone. The results generally support the hypothesis. Moreover, the pictures furnish a clear image of the deformation zone's growth patterns during loading, and this provides a critical test for future modelling efforts.

KEY WORDS Fracture Mechanics; rubber-toughened epoxy resins; thickness of adhesive layer.

INTRODUCTION

Structural adhesives are being used increasingly in extremely demanding applications and, as a result, there is a need to gain a better understanding of all aspects of their behaviour, particularly their fracture properties. In use, adhesives are subjected to a wide range of complex loads, so obviously a comprehensive understanding of their behaviour must include general loading. To develop this understanding, however, many programs, including this effort, have chosen to begin by studying what is arguably the simplest case, *i.e.* mode-I fracture. The acquired knowledge base must then be expanded to general loading, but it is hoped that principles can be developed in working with the simplest case that will be useful when moving to more complex loading and failure modes.

THEORY

From the many studies that have focussed on mode-I adhesive fracture, a great deal has been learned. For example, it has been established that for very brittle adhesives, the mode-I fracture surface energy, G_{IC}, for a bonded joint is essentially equal to the G_{IC} obtained for bulk samples of the adhesive material as long as the bonding surfaces are properly prepared to avoid interfacial failure.[1] When tough adhesives are used, however, the behaviour can be substantially different because the G_{IC} of the bonded joint depends on the thickness of the bondline.[2,3]

This is illustrated in Figure 1 where data are presented for adhesive G_{IC} as a function of bond thickness, T, for a model brittle adhesive and a model tough adhesive.[2] The brittle material exhibits no bond thickness effect and has a G_{IC} equal to that measured for bulk samples. For the tough material, however, bulk samples of the adhesive material gave a G_{IC} equivalent to that of the plateau found at high bond thicknesses, and hence the bonded joint can have G_{IC} values greater than or less than that of the bulk material depending on what bond thickness is tested.[4]

An understanding of adhesive fracture clearly requires an explanation for this bond thickness effect. Work over the past 10 years has developed an hypothesis which is at least consistent with the experimental observations. This explanation was developed in the work of Bascom, Wang, Hunston and Kinloch[2-5] and considers the combination of two effects. Both effects arise from the observation

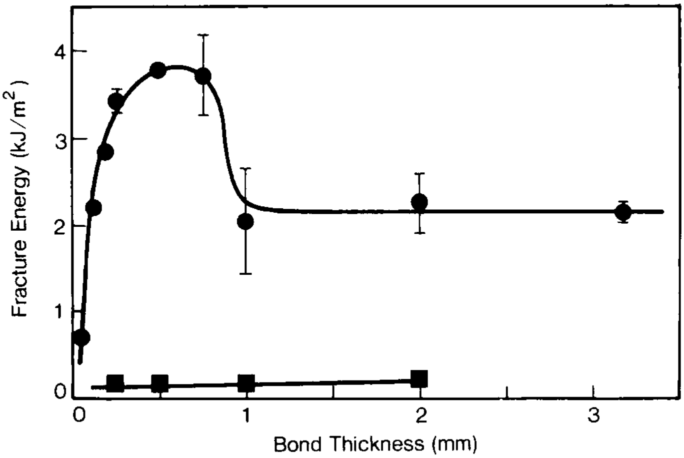


FIGURE 1 Adhesive fracture surface energy *vs.* bond thickness for an epoxy (■) and a rubber-toughened epoxy (●) from Ref. 2.

that tough materials owe their high fracture surface energies to their ability to generate crack-tip deformation zones of significant size prior to fracture. The deformation zone may be viewed as a mechanism to suppress the onset of rapid crack growth but, as the loading increases, the size of the zone must also grow to maintain its effectiveness. The toughness of the material depends on how large the zone can grow before the crack becomes unstable and rapid crack growth initiates. This depends on test conditions such as temperature and loading rate, but in most cases there is a good correlation between the size (or volume) of the zone ahead of the crack tip at the time of failure and the fracture surface energy.[6,7]

The adhesive bond hypothesis asserts that two other factors act to augment and limit the zone size and therefore alter the fracture surface energy accordingly. The first factor[2,3] is the limitation imposed on the zone height, D_h, by the adherends as the bond thickness, T, decreases (see Figure 2). The basis for the proposal is that D_h cannot exceed T, and consequently, as T decreases, the rigid adherends force a corresponding reduction in D_h. This leads to the prediction that the effect would become a major factor when T is equal to or less than the value of D_h in a corresponding bulk sample, D_{hB}, where there is no constraint. The resulting assertion is that the adhesive G_{IC} would decrease as the bond thickness is decreased below D_{hB}.

The second factor[4,5] that was proposed to alter the zone size involves the crack-tip stress field in the adhesive joint. Figure 3 shows a schematic for the tensile component of the stress ahead of the crack tip for different bond thicknesses. This prediction is taken from a linear elastic finite element analysis by Wang, Mandell, and McGarry.[8] In a very thick bond a stress distribution would naturally be expected to approach that found in a bulk specimen of the adhesive. Consequently, thick adhesive bonds and bulk samples of the adhesive are predicted to have similar behaviour, and this is exactly what is observed experimentally.[4] When the bond thickness decreases, however, the analysis indicated that a shoulder developed on the stress distribution with the result that

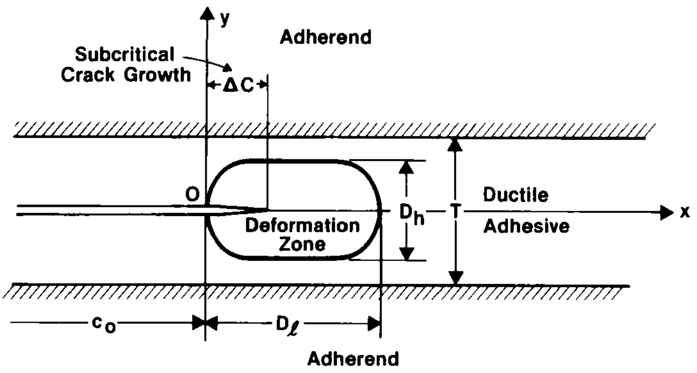


FIGURE 2 Schematic of crack-tip deformation zone. The crack length prior to application of load is c_0.

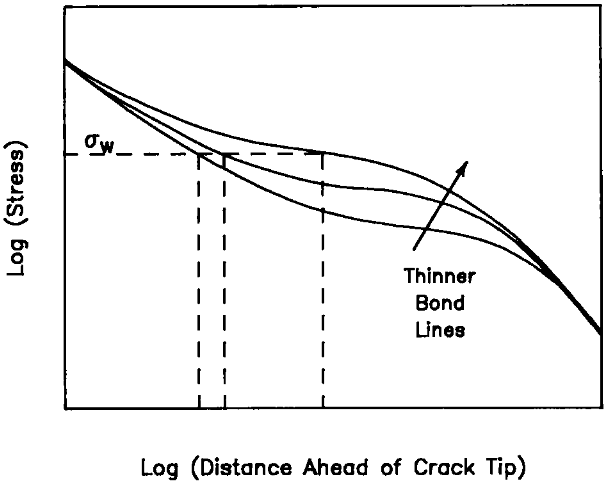


FIGURE 3 Schematic of crack-tip stress field (tensile component) for three different bond thicknesses. The proposed minimum stress for the formation of the stress whitening or deformation zone is σ_w (Ref. 8).

higher than expected stresses extended down the bond line ahead of the crack tip. If the minimum stress for the onset of the deformation zone is designated σ_w, Figure 3 would predict that the deformation zone would extend further down the bond line in thin joints than it does in a thick bond; *i.e.* the zone length, D_l in Figure 2, would increase. If D_l increases and D_h does not change or decreases slowly, the zone size and hence the fracture toughness should increase. Consequently, the fracture surface energy would be greater than that in the bulk sample. Logically, this effect should be most pronounced for bond thicknesses greater than D_h, but, of course, at high bond thicknesses the adhesive fracture behaviour must reduce to that characteristic of bulk samples.

The hypothesis proposed to explain the bond thickness effect asserts that both effects are present. Neither contributes in very thick bonds so the behaviour is equivalent to bulk samples. As the bond thickness is decreased, the size of the zone at failure begins to grow through increases in D_l produced by the stress field effect. Hence the fracture surface energy increases. Eventually, the bond thickness decreases to the point where it is roughly equal to D_h. Beyond this value, the constraining effect of the adherends becomes predominant, and thus the zone size decreases even though D may continue to increase. This produces a decrease in the fracture surface energy. The maximum in the adhesive bond G_{IC} is proposed to occur at bond thicknesses near D_h. For this simple explanation to be appropriate, D_h must not exhibit large variations with changes in bond thickness and should be approximately equal to D_{hB}. The temperature and loading rate must also be equivalent for all tests.

This explanation is obviously over-simplified since it ignores details which are known to be important. For example, the fracture surface energy depends not only on the size of the deformation zone but on other factors such as the stress–strain field present in the zone. Nevertheless, the hypothesis is qualitatively consistent with the measured fracture behaviour (Figure 1), and thus provides a basis on which to construct more complex models and a framework in which alternative explanations can be evaluated.

In this connection it is important to conduct additional theoretical and experimental examinations of the bond thickness effect to test, refine, or alter the ideas in the hypothesis. During the last few years a number of such studies have been performed. First, a detailed stress analysis was conducted and, as indicated previously,[5] it qualitatively supports the hypothesis described above although some details differ. A complete report on this analysis will be given elsewhere. Theoretical studies at NASA Langley Research Center[9] have also provided some support for the general picture described above. In addition, direct observations of the crack tip deformation zone were conducted using motion pictures. The results of these experiments are again consistent with the hypothesis. Although this conclusion has been reported in previous publications,[1,5] the experimental results have not yet been reported in detail. The purpose of this paper is to provide such a report.

EXPERIMENTAL

Fracture experiments were performed on bulk and adhesive specimens made from a model adhesive. It is composed of an epoxy (diglycidyl ether of bisphenol A cured with 5 parts of piperidine per hundred parts of epoxy (5 pphr). To toughen the system, 18.5 pphr of an elastomer (carboxyl-terminated polybutadiene acrylonitrile) were added to the epoxy-piperidine mixture prior to cure. The material was cured at 120°C for 16 hours.

The bulk samples were prepared in metal molds coated with a release agent, while the adhesive specimens were prepared with aluminium adherends that had been treated with a chromic acid etch to prepare the surfaces for bonding. Complete details on the samples and their preparation can be found in Refs. 1–3 and 6. This system was chosen because it exhibits stress whitening in regions of high load, and this whitening can be used as a relative measure of the deformation zone size and shape.

The fracture experiments were identical to those performed previously[1–3,10] and utilized the tapered double cantilever beam geometry shown in Figure 4. The specimens were loaded to failure in a tensile machine run at a constant cross-head speed of 0.02 mm/s and a temperature of 23°C. In all cases a point was reached in the loading where rapid crack growth occurred, and this point was defined as failure. A motion picture camera was used to monitor the crack tip region throughout the fracture test. The film speed was 50 frames per second. Simple lighting on the same side as the camera was found to be sufficient clearly to define the stress whitening at the crack tip. The film was then analyzed to determine the length, D_l, and height, D_h, of the stress whitened zone (see Figure 2). In some cases the crack was found to grow slowly prior to the onset of rapid crack growth.

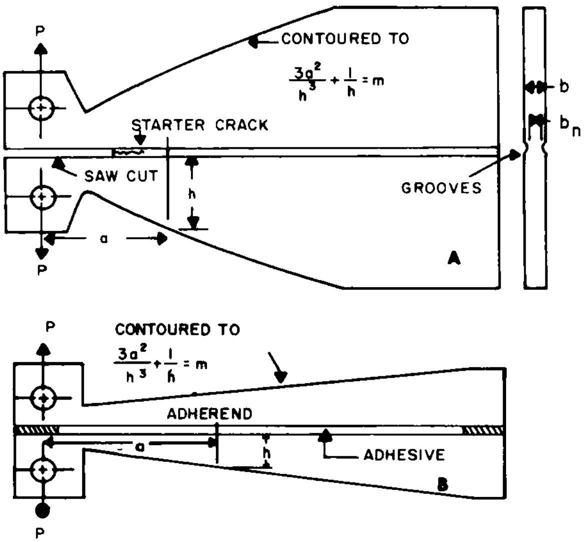


FIGURE 4 Diagram of tapered double cantilever beam specimen for bulk (A) and adhesive (B) tests. The taper, m, is: $3\,\text{cm}^{-1}$ for bulk samples and $90\,\text{cm}^{-1}$ for adhesive specimens. The sample thickness, b, is 1.25 cm with wide side grooves in the bulk specimens to help guide the crack growth.

Any such subcritical crack growth was measured as Δc. A critical assumption here, of course, is that the stress whitening provides at least a relative measure of the deformation zone.

Experiments were performed on bulk specimens and bonded joints with thicknesses of 0.38 mm, 1.0 mm, and 2.4 mm. The materials and test conditions used here are virtually identical to those used in the experiments which generated Figure 1. Consequently, the 0.38 mm bond is near the maximum in the bond thickness curve, while the 1.0 mm and 2.4 mm bonds are on the plateau associated with thick bonds. This means that comparisons of the zone size and shape in the different cases provides a direct test of the hypothesis presented above.

The 2.4 mm thickness is unrealistically large for a bonded joint, and thus only a few samples were tested. These particular samples produced excellent pictures but exhibited large values of G_{IC} that are not typical of those determined in a test where a sufficient number of specimens were used to obtain a truly representative value. The clear pictures obtained for these samples provide excellent data for a detailed analysis of zone growth, and a future publication will report such an examination and comparisons with the predictions of finite element analyses. In this paper, however, the objective is to examine the relative size and shape of the zone as a function of bond thickness, and thus the results used must be typical for the various thicknesses. Consequently, the results for the 2.4 mm bonds were not used in the comparisons here.

RESULTS

The results in the movies can be illustrated with Figure 5 which shows a still from the sequence for a 2.4 mm bond. This photograph is less than 0.02 seconds from the onset of rapid crack growth. The stress whitening is clearly visible. During loading this stress whitening is first observed as a small zone ahead of the crack tip at a load that is 20% to 30% of the failure load. Perhaps by coincidence the equivalent unmodified epoxy failed at about 20% of the failure load for this toughened specimen. Figure 5 also illustrates an important feature of the failure behaviour. The stress whitening is always generated ahead of the crack tip. In Figure 5 the initial precrack ended at the extreme left of the photograph. Prior to the onset of rapid crack growth, however, the 2.4 mm bond exhibited significant slow (subcritical) crack growth. As seen in Figure 5, the crack tip has advanced to a point well within the stress whitened zone when the failure load is achieved. Although the left side of the stress whitened region is unloaded as the crack tip advances past it, the material remains stress whitened. In general where subcritical crack growth is observed (2.4 mm bonds and bulk specimens), it occurs during the last third of the loading.

Examples of typical results for bonds with thicknesses of 0.38 mm and 1.0 mm are shown in Figures 6 and 7. These Figures plot measurements of D_h and D_l as a function of time during loading. To facilitate comparisons, the times are normalized by the time to failure for each specimen. No subcritical crack growth was seen for bonds of either thickness, and so Δc was 0. The results shown in these Figures are typical of the data for those bond thicknesses.

The bulk specimens were tested in only a limited number of experiments, but

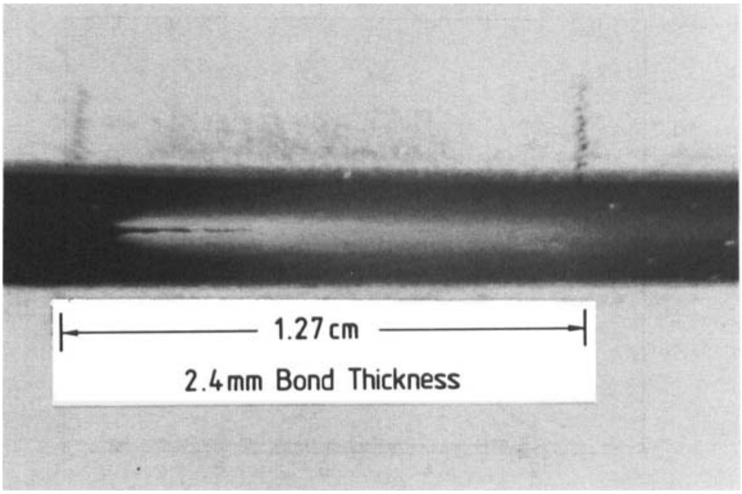


FIGURE 5 Picture from movie sequence on fracture of 2.4 mm adhesive joint taken just prior to failure. See color Plate I.

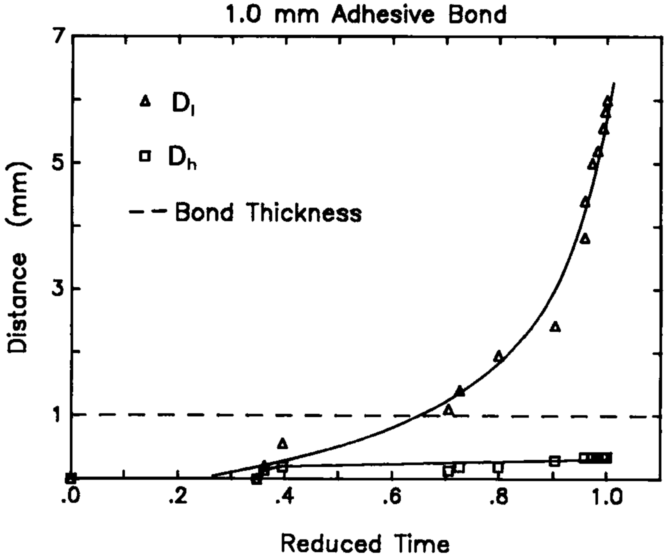


FIGURE 6 Measurements of deformation zone growth in a 1.0 mm bond as a function of reduced time (time divided by the time at failure, 28.8 s).

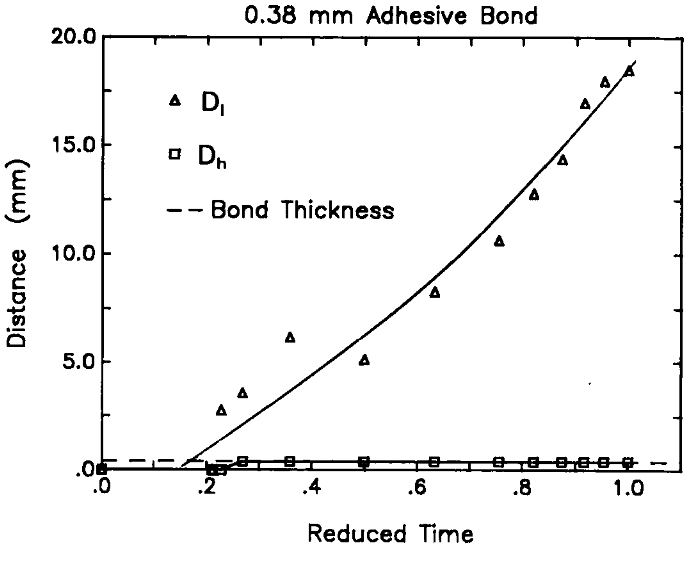


FIGURE 7 Measurements of deformation zone growth in a 0.38 mm bond as a function of reduced time (time divided by the time at failure, 16.3 s).

the results show some important similarities and differences relative to the behaviour of the thick adhesive bonds (i.e. the 1.0 mm results). The length of the zone, D_l, is slightly smaller in bulk samples than in 1.0 mm bonds. Moreover, bulk specimens exhibit subcritical crack growth for the last third of the loading. By the time the failure point is reached, Δc is 20% or more of D_l. Consequently, the length of the zone ahead of the crack tip is substantially less than in the 1.0 mm bonded joints. On the other hand, D_h in the bulk samples is significantly greater than in the 1.0 mm bonds. This means the bulk samples produce a zone that is more circular than the highly elliptical zone found in the adhesive bonds. For the limited number of specimens measured here, however, the volumes of the zones ahead of the crack tips are quite similar for bulk specimens and 1.0 mm bonds.

DISCUSSION

Based on results such as those in Figures 6 and 7, four observations can be made. The first two provide a test of the bond thickness hypothesis. Observation one concerns the size of the deformation zone at failure, and how it changes with bond thickness variations. In going from bulk samples to thick adhesive bonds the volume of the zone changes relatively little, but a change in shape is observed. This is contrary to the picture offered in the bond thickness hypothesis and suggests that the real situation is more complex than the simple ideas presented above. Nevertheless, the conclusion remains the same since the zone volume does not change greatly, and this is proposed to the critical parameter. Such a result is not surprising since the fracture surface energies of these specimens are quite similar. As the bond thickness continues to decrease (data for 0.38 mm bond), however, D_l becomes significantly larger as can be seen in Figures 7 and 8. Figure 8 shows pictures taken from the movies using the last frames prior to failure by rapid crack growth for a 0.38 mm bond. The difference in D_l can clearly be seen as well as the absence of any subcritical crack growth. Figure 8 shows that the zone size increased as the thickness was reduced from 1.0 mm to 0.38 mm, and this provides an explanation for the increased fracture surface energy. This is exactly what the bond thickness hypothesis predicts.

The second observation that can be made from the motion pictures concerns the comparison between the height of the zone and the bond thickness. Experimental observations show that, unlike the simple hypothesis, the bulk samples exhibit larger values of D_h than the bonded joints. On the other hand, Figures 6 and 7 show that D_h does not differ greatly for the two adhesive geometries tested. Moreover, for the 0.38 mm bond (Figure 7) D_h is equal to the bond thickness. This is in agreement with the hypothesis which suggests that D_h should equal T at the maximum adhesive G_{IC}. It also follows that bond thicknesses less than 0.38 mm will have a reduced zone height. This does not prove that the zone size will be smaller since D_l may also be increasing, but it is consistent with the hypothesis. Thinner bonds could be used to test this idea, but

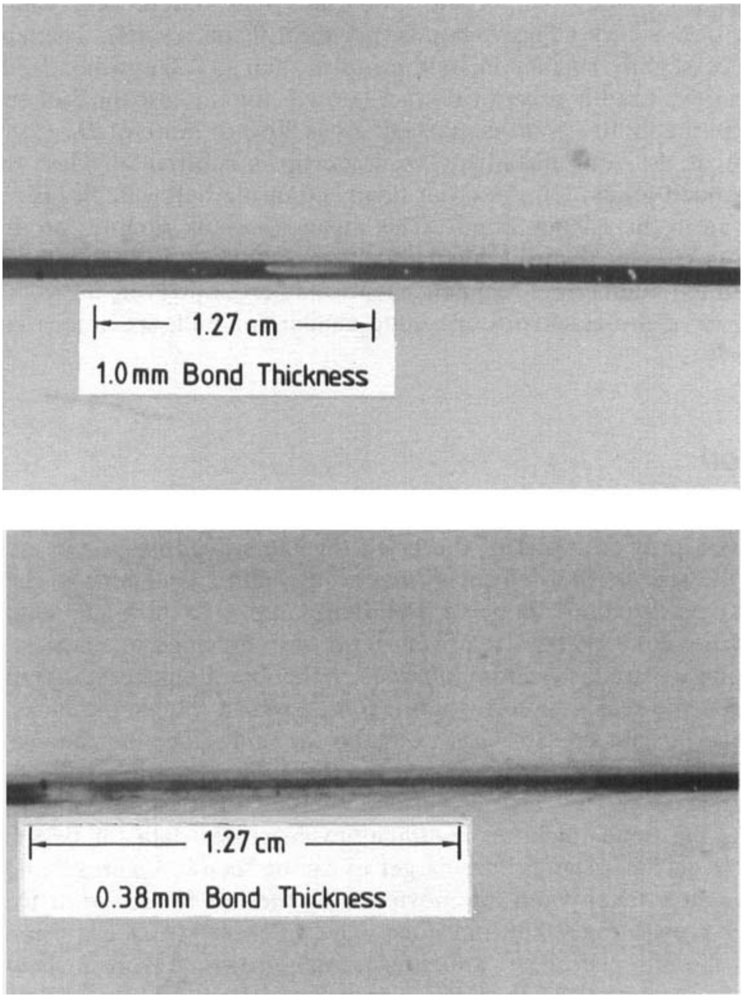


FIGURE 8 Picture of deformation zone just prior to failure for two different bond thicknesses. See color Plate II.

unfortunately, they are difficult to see in the pictures. Post failure observations of the fracture surface to estimate D_l are not completely reliable because some of the stress whitening may be lost when the load is removed. Consequently, it is possible only to say that the results obtained here are consistent with the hypothesis for very thin bonds.

The third observation that can be made concerns differences in behaviour for adhesive bonds and bulk samples. As indicated previously, the bulk samples displayed a less elliptical zone shape than the bonded joints and exhibited subcritical crack growth prior to fracture. Similar subcritical crack growth was seen for 2.4 mm bonds. On the other hand, neither the 0.38 mm bonds nor the

1.0 mm bonds gave any significant subcritical crack growth. The former are at the peak of the bond thickness curve and therefore might be expected to show different behaviour, but the latter are in the plateau region of Figure 1. Clearly, not all the bond thicknesses in the plateau exhibit the same behaviour or behaviour equivalent to bulk samples. Perhaps this is because the 1.0 mm bonds are at the extreme left end of the plateau, but it may also reflect the fact that the real situation is more complex than the simplistic hypothesis offered to explain the bond thickness effect.

The final observation that can be made concerns the growth patterns exhibited by the deformation zone. As illustrated in Figures 6 and 7, the zone is first detected about 1/3 of the way through the loading. The height, D_h, grows quickly and then levels off at a relatively constant value for the remainder of the loading even when it is not constrained by the bond thickness as it is in the 0.38 mm adhesive joint (Figure 7). The deformation zone length, D_l, however, exhibits an exponential growth as shown in Figure 6. For those geometries where subcritical crack growth is observed, Δc also exhibits an exponential growth pattern. Consequently, at the failure point, the growth rate for D_l (and Δc when present) is very high. The one possible exception is the 0.38 mm bond where the growth of D_l seems to be relatively constant throughout the loading although the data scatter makes quantitative comparisons difficult. Perhaps, in a thin bond, the geometry somehow suppresses the unstable nature of the usual zone growth pattern. If so, this may play a role in the ability of these bonds to support a larger zone and hence generate a higher fracture surface energy. In any case, not only is the size and shape of the zone different in thin bonds but the growth pattern may also be different.

CONCLUSIONS

The experiments conducted here show that the size of the crack-tip deformation zone at failure in an adhesive bond changes with alterations in bond thickness, and that these changes correlate with the measured variations in the fracture surface energy. In thick bonds the zone volume and fracture behaviour are similar to those found with bulk samples of the adhesive. As the bond thickness is decreased the zone size at failure grows by extending further down the bondline. The fracture surface energy shows a corresponding increase. Eventually a point is reached where further decreases in bond thickness cause the fracture surface energy to decrease. This change occurs at approximately the point where the height of the deformation zone is equal to the bond thickness.

Observations of zone growth also show that bulk samples and very thick bonds exhibit subcritical crack growth prior to failure while the thinner adhesive bonds show little evidence of this growth. With the possible exception of the thinnest bonds tested, the length of the deformation zone and, where present, the subcritical crack growth show an exponential growth pattern resulting in very high rates of growth just prior to failure. For the thinnest bond the length of the zone seems to exhibit a more stable growth pattern, but the data scatter makes it difficult to quantify this difference.

References

1. D. L. Hunston, and W. D. Bascom, *Rubber-Modified Thermoset Resins, Advances in Chemistry Ser. 208,* C. K. Riew, and J. K. Gillham, Eds., (American Chemical Society, 1983), Chap. 7.
2. W. D. Bascom, R. L. Cottington, R. L. Jones, and P. Peyser, *J. Appl. Polym. Sci.* **19,** 2545 (1975).
3. W. D. Bascom and R. L. Cottington, *J. Adhesion,* **7,** 333 (1976).
4. A. J. Kinloch and S. J. Shaw, *J. Adhesion,* **12,** 59 (1981).
5. D. L. Hunston, A. J. Kinloch, S. J. Shaw, and S. S. Wang, *Adhesive Joints: Formation, Characterization, and Testing,* Mittal, K. L., Ed. (Plenum Press, 1982), pp. 789–808.
6. A. J. Kinloch, S. J. Shaw, D. A. Tod, and D. L. Hunston, *Polymer,* **23,** 1341 (1983).
7. A. J. Kinloch, S. J. Shaw, and D. L. Hunston, *Polymer,* **24,** 1355 (1983).
8. S. S. Wang, J. F. Mandell, and F. J. McGarry, *International Journal of Fracture,* **14,** 39 (1978).
9. J. H. Crews, K. N. Shivakumar, and I. S. Raju, *NASA Technical Memorandum 89033,* (NASA Langley Research Center), November, 1986.
10. S. Mostovoy, and E. J. Ripling, *J. Appl. Polym. Sci.* **10,** 1351 (1966).