The Constitution of an Uncrosslinked Polymer in the Process of Damage Accumulation

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ABSTRACT

Current thinking in nonlinear fracture mechanics research is closely coupled to the behavior of material behavior at the tip of a crack; it is often associated with the development of voids that grow before total material disassociation occurs, their coalescence being considered synonymous with fracture. We describe experiments which provide a measure of the stress-strain behavior of material in the process of developing such void-damage. The stress and strain measures are suitably averaged over a domain on the order of 0.1mm which is large compared to the size of the voids. As a model material we use Polyvinylacetate, an uncrosslinked (thermoplastic) polymer, having a glass transition temperature near 30°C. The experimental vehicle is a bonded DCB specimen subjected to displacement determinations with the aid of optical interferometry.

KEYWORDS

Non-linear stress-strain behavior, fracture, void growth, polymer

INTRODUCTION

Fracture processes are usually associated with high intensity shear deformations accompanied by the development and growth of voids. The fracture progression occurs then usually through the coalescence of these voids. In order to model the fracture process it is necessary to determine the "properties" of the voided and failing material, for if these properties are known, it becomes possible to determine the fracture behavior of a solid without recourse to an independently postulated fracture criterion.

These observations are true for virtually all materials, though the impetus for the present investigation derived from the need to better understand the failure behavior of uncrosslinked polymers for use in advanced composite materials. In these latter materials the failure (initiation) behavior is dominated by the close proximity of the second material phase (fibers) and thus influences the development of the toughness through restricting the size of the domain in which the damage process takes place. When compared to thermosetting polymers the deformations in certain thermoplastic polymers are associated with larger amounts of energy dissipation before fracture/failure ensues. In order to capitalize on this feature it appears attractive to use thermoplastics in advanced composites. However, the large amount of energy dissipation accompanies a certain void-associated deformation mechanism which requires a minimum size scale in order to fully develop. Inasmuch as the space available to the matrix material between fibers is, at best, limited to dimensions on the order of a few microns it is of interest to investigate the failure response and development of the polymer under such strong spatial constraints. In the following development we retain, however, a more general outlook on the failure behavior of the test material.

The quantitative behavior by which material failure occurs through voiding is, at best, poorly understood. This is primarily so because the dimensions of the domains in which this mechanical breakdown occurs is (usually) extremely small, so that direct observation becomes very difficult. Accordingly, most of our information regarding this failure process derives from post mortem fractographic analyses. In order to cope with this type of microscopic failure process in terms of the macroscopic concepts underlying continuum mechanical analyses, it is necessary to cast the average properties of the failing or voiding material in continuum rather than microscopic descrete terms. The failing material needs to be represented, therefore, in terms of a nonlinear constitutive behavior on a size scale that is large compared to the detailed and microscopic features developing in the material.

To date there seems to have been only few serious attempts to address such material behavior experimentally. As noted above, the main reason for this lack of experimental effort is the usually small size scale of the domain in which such phenomena operate. However, under some conditions and with certain materials the size scale of this domain can be controlled to sufficiently large dimensions so as to allow the determination of such average properties, at least in some way and to a degree that possibly allows guidance to and corroboration of parallel analytical efforts.

An experimental situation that permits estimation of such behavior arises when the material of interest undergoes sufficient "plastic" deformation and flow and, when formed into a thin layer, and deformed normal to the layer plane between relatively rigid planes to which it can be attached or bonded securely without interfacial failure. The geometry for this purpose is shown in Fig. 1, which is basically a bonded double cantilever beam geometry often used in adhesion studies and simulates the geometry used for determining the fracture energy associated with composite delamination such as was employed in (Chai et al., 1983, Marceau et al., 1977).

However, while in those earlier studies the objective of the work was to elaborate on the usefulness or the determination of the energy release rate as a method of characterizing the fracture process, the present study has a different aim in mind. The underlying motive behind the present work is to provide initial experimental characterization of the fracture process on a more detailed level of material behavior than can be provided by the "averaged" characterization by a fracture or cohesive energy. While the work presented here does draw on certain approximations, these are believed to be commensurate with an initial attempt at exploring both the method of properties determination as well as the estimation of this type of nonlinear behavior.

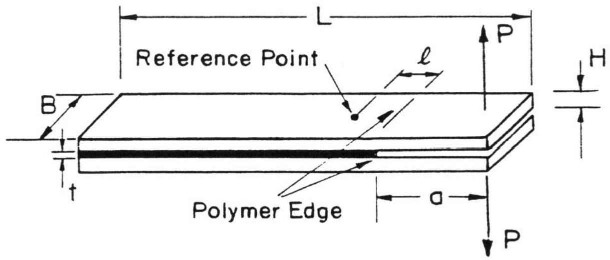

Figure 1. Test specimen geometry typical dimensions $H = 8$ mm, $t = 0.025$ mm, $B = 25$ mm, $L = 170$ mm, $a = 45$ mm,

The method of determining the desired constitutive behavior of the failing material rests on measuring the deformations of the metallic solids and on deducing from them the cohesive forces as the load is increased to the point where the material at the polymer front proceeds to failure. The analytical counterpart of this problem has been considered in detail in references by Ungsuwarungsri et al., (1987, 1988) including the effect of of nonlinear cohesive forces under exclusion of plastic deformations in the metal components. The problem can, in principle, be solved by determining the displacements and deformations of the metal components under increasing load, including the displacements on that part of the beam surface where the voiding material exerts tractions; the average constitutive behavior can then be determined from the tractions (reaction) offered by the voiding material as the solution to a standard, linear boundary value problem, along with the average strain across the layer thickness, computed from the measured surface displacements and the layer thickness.

In the sequel we discuss the suitability of the test geometry to this purpose and the experimental procedure. We then discuss the limitations imposed on the evaluation of the displacements by the joining of the different materials. In effect this limitation results in the substitution of a different boundary value problem for that desired optimally.

TEST GEOMETRY

The test geometry used (Fig. 1), is essentially a double cantilever beam (DCB). In conjunction with a relatively stiff testing machine the geometry allows generally stable damage growth. In order to obtain information on the behavior of the voiding material, two conditions must be met, namely that a) the test material can be made to adhere sufficiently well to the metal (aluminum) beams to allow failure away from the polymer/aluminum interface, and b) the deformations in the beam are so small as to invoke only elastic behavior. For the latter reason a beam thickness of 3/8 inch was found suitable. This choice required then a zone size over which the beam deformations were measured for the evaluation of the interface tractions which was about the same as the beam thickness.

Condition a) above was met by resorting to pressure bonding the polymer to the etched beam surfaces (optimized FPL etch process, see the references by Parvin et al., 1987). Specimens were manufactured by first (pressure) bonding two $H = 3/8$ inch thick plates of 2024T3 and 6inch x 6inch in laterial dimensions. The surface of one of these bonded plates was then lapped optically flat to within about 3 wavelengths of light per inch and then polished to a mirror finish. DCB specimens with the dimensions shown in the caption to Fig. 1 were then cut from these plates and prepared for further adaptation to the loading device for attachment to an Instron Tester. For convenience in data analysis the specimen was also held in a special mechanism that restricted the net rotation of the specimen during load application.

EXPERIMENTAL PROCEDURE

Surface deflections of the specimen under different loads were determined with a Michelson interferometer as illustrated in Fig. 2. In order to obtain an absolute measure of the beam deformations it would be desirable to make simultaneous measurements over the entire surface of the specimen including the region of zero displacements. The field of view of the interferometer precluded such a large observation domain and only a portion of the displacement field could be determined. In addition, it was necessary to content with small rotations that did occur inspite of the levelling device which connected the specimen to the Instron Tester.

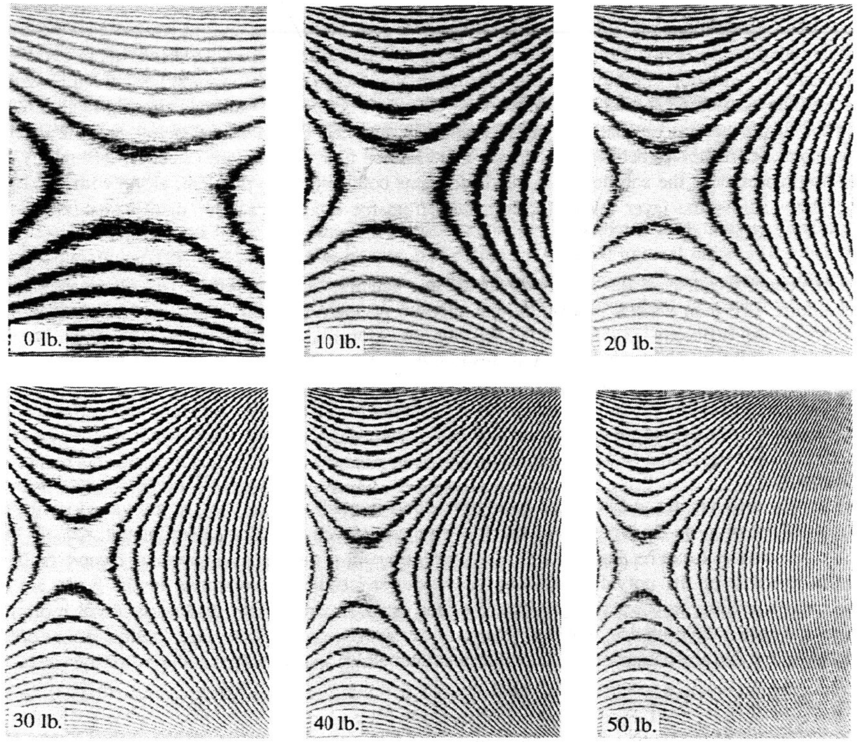

Figure 2. Interferograms of the specimen surface deformed under end loads as indicated.

These inevitably occurring, small rotations were accounted for by adjusting the mirror M2 (cf Fig. 3) and monitoring its rotation, which amounted to rigid body rotation. It should be recalled in this connection that rigid body rotations do not affect the tractions we seek. The only context in which these rigid body rotations must be accounted for is in the computation of the average strain across the polymer layer, which is determined simply from the separation of the beam surfaces; only one-sided measurements are carried out and symmetry was assumed.

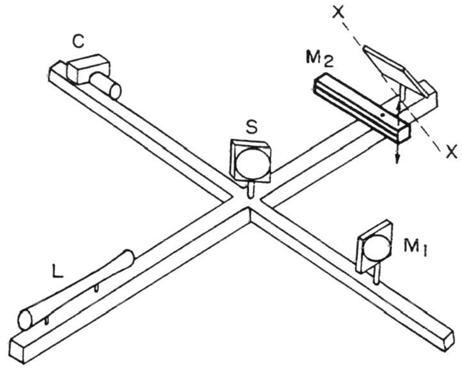

Figure 3. Michelson interferometer, schematic. Mirror M2 is the specimen.

We note in passing that even though the specimens were cut from plates that had been lapped optically flat, the DCB specimens exhibited deformations after being cut from the plates, (cf Fig. 2). deformations are consistent with those of the "opening mode" type and are tentatively interpreted as resulting from residual stresses incurred in the specimen manufacturing process. Inasmuch as these deformations are easily recorded they may be subtracted from the deformations recorded under loading. One may question, of course, whether these initial deformations should be eliminated from the determination of the cohesive tractions in the polymer. In response we can only state that a) we have no direct knowledge of the magnitude of these stresses, and b) they are considerably lower than the deformation state might indicate because of the (viscoelastic) relaxation that occured between the time of specimen preparation and start of the experiments (days). In summary, we do not believe that we incur a major error in omitting these stresses from consideration, yet such considerations must surely be part of a future and more refined study.

LOAD HISTORY

Since PVAc possesses a glass transition temperature of about 30°C, any deformation at room temperature will ellicit some viscoelastic response. In order to minimize variations due to that effect [1] only a single load history was employed: Measurements resulted from a nearly constant displacement rate of the beam ends (0.5 mm/min; 0,02 in./min.). This deformation was interrupted only briefly at various load levels for mirror adjustments prior to data recording as described above under "Experimental Procedure". The time required for this adjustment and for the recording was about one minute. While load relaxation occured during these intervals, no comparative change in fringe

1. In this initial investigation it seemed unreasonably difficult to include effects resulting explicitly from the time dependence of the polymeric material.

spacing was observed and this relaxation was accounted for by consistently using the partially relaxed load value in the determination of the polymer induced tractions.

EXPERIMENTAL RESULTS

The information from which the stress-strain behavior of the voiding polymer is deduced arises in the form of fringe-displacement contours as exemplified in Fig. 2 and by the force-displacement trace deduced from conditions at the ends of the beams.

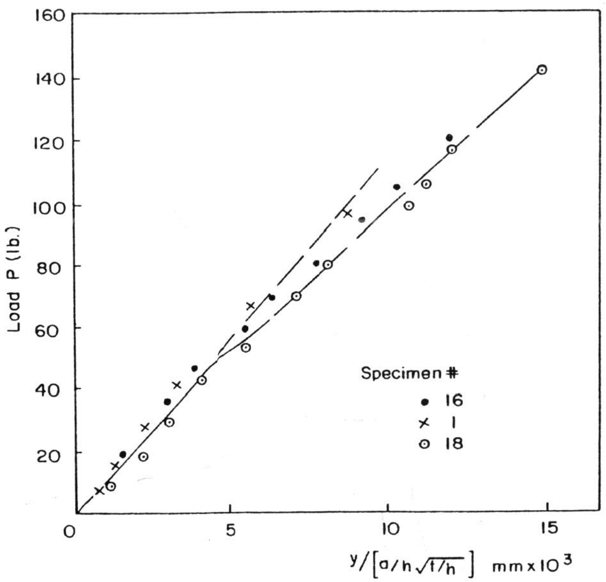

Figure 4. One half of "crack-opening displacement" as a function of beam end load.

Fig. 4 illustrates the relation between the force(s) applied to the end(s) of the specimen and the displacement across the polymer layer at the polymer edge (crack tip). This data was derived from specimens having different dimensions and the abscissa includes normalization consistent with parameters that describe the deformation of a beam on a Winkler foundation. It is clear that damage develops above the 40 to 45 lb. load range.

EVIDENCE OF VOID FORMATION

Examination of the exposed edge of the polymer layer along the side of the specimen demonstrated the development and growth of voids. While this process could be monitored during loading it was not feasible in our experimental arrangement to record this process through a microscope during the deformation process concurrently. Therefore, when void formation occurred, loading on the specimen was discontinued and a wedge was inserted to maintain the deflections of the beam ends. After

removal from the testing machine this deformed assembly could then be examined and photographed under the microscope. In this manner Fig. 5 of the voids were obtained.

It is of interest to note that the damage process seems to consist of void growth and coalescence at the front of the damage zone. Once these voids become sufficiently large (cf Fig. 5b) the connecting ligaments break over a discernable distance and a fracture advances by a discrete step, coming to rest with the tip in a region of smaller voids as shown in Fig. 5c.

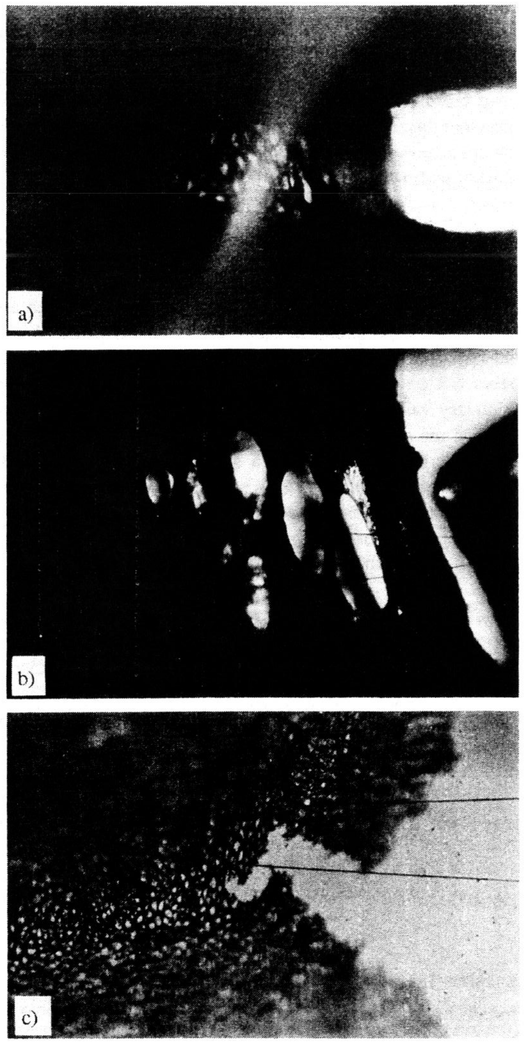

Figure 5. Void formation at the front of the polymer (crack-tip)

a) initial growth

b) coalescence and large voids

c) fracture propagation by discontinuous growth into void domain

We include here another observation related to the void size and failure under constrained deformation as it relates to the failure of composites and (uncrosslinked) adhesives. Note from Fig. 5c that there are many voids across the polymer thickness (though not necessarily uniformly so as assumed for data reduction later on) which are estimated to be on the order of 5 x 10^{-3}mm in the present state of deformation; in their initial formation state the voids would be smaller. On the basis of these dimensions it would be reasonable to expect that the same deformation and voiding process would occur in a layer of smaller thickness than studied here. However, the energy required to disintegrate or fracture the layer would depend on the layer thickness in two ways: First, the thinner the layer is, the fewer voids can be generated so that in this regard the failure energy should be (roughly) proportional to the layer thickness. Second, the thinner the layer and the fewer voids there are, the less interaction and less coalescence can take place. One would expect, therefore, that the voided material can sustain a smaller strain in a thin film before failure occurs than would be the case in a thicker one. For these two reasons one should expect a lower energy expenditure in the fracture process the thinner the polymer layer is. These considerations would seem to cease being valid when the layer is so thick that the voiding/failure process cannot be reasonably considered to occur uniformly across its thickness dimension.

EVALUATION OF THE MEASUREMENTS

Returning to the overall aim of this investigation we recall that the present considerations are to cover a size scale that averages over several (many) voids. In this way we may consider the voiding material to possess the average mechanical properties we seek here. In principle this response could be determined if the displacements OR tractions are determined on all surfaces of the beam elements, for then the solution to a boundary value problem formulated for the linearly elastic and homogeneous aluminum beams would yield the desired answer. We do not know the deformations at the interface(s) between polymer and aluminum [2]. However, a detailed finite element analysis (Ungswarungsri et al, 1987) showed that if the stiffness of the polymer is smaller by a factor of ten than the stiffness of the beams then the surface-normal displacements at the traction free surfaces were equal to those at the interface with a deviation of maximally 4%. Therefore, we shall use the displacements determined on the exposed surface of the beam in place of the interfacial ones.

It turns out that in spite of the high precision with which the displacements are determined the solution of the appropriate boundary value problem leads to tractions over the surface domain where the displacements are determined which oscillate as a function of the number of points where displacements are determined. These oscillations are clearly the effect of small inaccuracies in the displacement field. To eliminate these oscillations it was desirable to smooth the data through fitting a polynomial to it by least squares. It must then be recognized that such a smoothing operation also restricts the type of solution that results: Combining this operation with the use of the surface displacements instead of the interfacial ones is equivalent to treating the aluminum adherends as (Timoschenko) beams. For this reason beam theory was used to estimate the cohesive tractions offered by the polymer.

From Fig. 2 it is evident that the beam surfaces deform with anticlastic curvature; we shall not be

2. Attempts at replacing the aluminum beams with glass ones for accessing the interface deformations directly failed, either because the glass broke in the test before polymer failure occured, or because the bond between glass and polymer could not be made sufficiently strong.

concerned with the deformations of the whole beam but concentrate only on those along its centerline. A least square fit of the polynomial

$$y = \sum_{n=0}^{N} a_n x^n \tag{1}$$

to the experimental data permits the computation of the moment, shear and distributed loading on the beam. While the polynomial fit should determine the appropriate constants from which the loading on the beam can be deduced, it is necessary to also ensure that at least the moment and shear boundary conditions acting at the "crack tip" are satisfied. The boundary conditions in the interior region are less stringent since they address slope and displacement conditions which are automatically included in the measurements. Some experimentation with polynomials of different degrees allowed for sixth order with larger degrees rendering little advantage inasmuch as the number of points available (the number of fringes in the region of interest) for a least squares fit was also limited to about 10 for the lower load levels, and 30 to 40 for the longest, loads [3]. A sixth order polynomial was found to represent the data quite adequately [4].

The loading was thus determined from the beam equation

$$\frac{q}{EI} = y'''' = \sum_{n=4}^{6} n\,(n-1)\,(n-2)\,(n-3)\,a_n\,x^{n-4} \tag{2}$$

subject to the conditions at the "crack tip"

$$\frac{M}{EI} = y'' = \sum_{n=2}^{6} n\,(n-1)\,a_n\,x^{n-2} = \frac{Pa}{EI} \tag{3}$$

$$\frac{V}{EI} = y''' = \sum_{n=3}^{6} n\,(n-1)\,(n-2)\,a_n\,x^{n-3} = \frac{P}{EI} \tag{4}$$

The loading q was determined in this way as a function of position along the interface. Inasmuch as the displacement is also known at each point along the interface the average strain across the polymer layer is determined as a function of the coordinate along the interface and the cohesive forces can be determined as a function of this average strain. Such data has been compiled in Fig. 6 for displacement measurements corresponding to several load levels. With increasing loading the maximum strain achieved (at the crack tip) increases also: the agreement or consistency of the "stress-strain" behavior at the lower load levels with those at higher ones is a measure of the quality of the data and, to some extent, a confirmation of the adequacy of the data reduction scheme.

3. Increasing the polynomial degree allowed resurgence of the non-smooth behavior of the computed tractions (reaction loading).

4. A computation based on Timoshenko beam theory was also accomplished. It turned out to yield results virtually indistinguishable from those reported here.

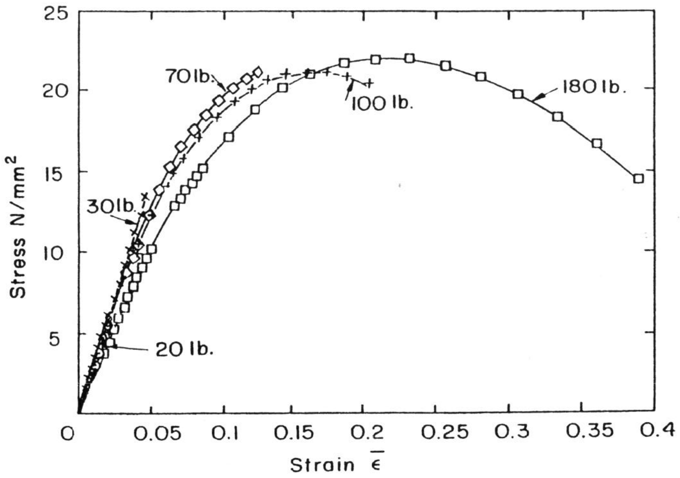

Figure 6. Stress q as a function of the average strain across the thickness of the polymer layer.

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