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The measurement of damage initiation, particle adhesion and cohesive strength from traction displacement curves of a nano-toughened epoxy

D. Mc Auliffe [a], A. Karac [b], N. Murphy [a], A. Ivankovic [a,*]

[a] School of Mechanical and Materials Engineering, University College Dublin, Belfield, Dublin 4, Ireland
[b] Masinski fakultet u Zenici, Fakultetska 1, 72000 Zenica, Bosnia and Herzegovina

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ABSTRACT

In this work the deformation behaviour of a nano-toughened epoxy adhesive is measured at different levels of stress triaxiality. The test method consists of a notched axisymmetric adhesive layer loaded in tension. The recorded traction displacement curves were analysed numerically and it was found that the measured peak stress corresponds to the intrinsic cohesive strength, σ_{max} of the material. This method allows experimental measurement of σ_{max} for use in cohesive zone models of fracture. Additional features of the traction displacement curves include a kink that corresponds to particle debonding at a critical hydrostatic stress. By application of the Mori–Tanaka model, the relationship between the experimental measurements and particle/matrix adhesion is described.

1. Introduction

Epoxy based structural adhesives have been toughened by second phase particles for many years. More recently, nano-sized particles have been added to epoxies to produce adhesives with fracture energies over an order of magnitude greater than their epoxy matrix alone. For nano-toughened adhesives, this increase in fracture energy is derived from localised plasticity that occurs due to the presence of the particles. The two primary mechanisms are growth of voids initiated by particle debonding or cavitation, and plastic shear bands which develop between particles/voids [1–5].

Adhesive joint fracture is often modelled using a cohesive zone model (CZM) [6–8]. The CZM is governed by a traction separation law (TSL), which relates the separation of opposing fracture surfaces to the traction across them. This approach is preferred to modelling the mechanisms of material separation directly since the length scales involved differ by several orders of magnitude. The critical parameters of a traction separation law are the intrinsic fracture energy G_0 and the maximum cohesive strength σ_{max}. If the CZM is coupled with an elastic plastic continuum the total fracture energy G_C is the sum of energy dissipated in the CZM, G_0, and the plastic energy dissipated in the continuum, G_p, i.e.

$$G_C = G_0 + G_P \quad (1)$$

Tvergaard and Hutchinson [9] noted that the ratio of the σ_{max} to the yield stress σ_y must be greater than 2.5 for G_P to be significant compared to the G_0. The cohesive strength is often assumed to be greater than the strength of the material under uniaxial tension due to the constraint effects during fracture. Pardoen et al. [6] used a CZM described by a single TSL (i.e. constant G_0 and σ_{max}) with $\sigma_{max}/\sigma_y > 3$ coupled to an elastoplastic adhesive layer to model the fracture of adhesive joints of different thickness and obtained close agreement. Cooper et al. [10] modelled the fracture behaviour of tapered double cantilever beam (TDCB) tests conducted over a range of bond gap thicknesses using a variable TSL, with $\sigma_{max}/\sigma_y < 2$ in all cases and found that G_0 accounted for most of the energy dissipation.

Another approach [11] employs critical distance theory whereby fracture occurs when a failure stress at a critical distance is reached. The critical distance and failure stress were determined by inverse analysis, and it was demonstrated that the critical distance related to microstructural features. With this model $\sigma_{max}/\sigma_y > 3$ was used.

Clearly the details of energy dissipation and interpretation of numerical analyses are highly dependent on the set of cohesive parameters chosen to simulate fracture. Studies regularly rely on inverse analysis to determine the value for σ_{max} and G_0 [12,6] whereby the parameters are varied until satisfactory agreement is obtained. Indeed it has been shown that multiple combinations of

* Corresponding author.
E-mail address: Alojz.ivankovic@ucd.ie (A. Ivankovic).

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σ_{max} and G_0 produced agreement with experimental results [13,10]. Therefore, direct experimental measurement of σ_{max} is desirable to define a TSL.

The fracture behaviour of most materials is sensitive to the degree of stress triaxiality at the crack tip. High stress triaxiality suppresses yielding which encourages alternative fracture processes to occur. The mechanisms of fracture for nano-toughened adhesives are primarily shear banding and plastic void growth and their development is affected by the stress triaxiality [14,2,15]. Therefore it is useful to characterise material behaviour under conditions of high stress triaxiality. A common measure of stress triaxiality, H, is given as:

$$H = \frac{\sigma_{hyd}}{\sigma_{vm}} \tag{2}$$

where σ_{hyd} is the hydrostatic stress and σ_{vm} is the von Mises stress.

Some studies have investigated the effects of stress triaxiality on the deformation behaviour of bulk materials using a tensile specimen containing an axisymmetric notch. Deformation is localised to the notched region due to the reduced cross-sectional area, but the stress state also becomes triaxial across the remaining ligament. The degree of stress triaxiality is dependent on the depth and geometry of the notch as well as the elastic constants of the material [10,16–18]. Pardoen et al. [19] studied void growth in copper bars using this technique. H was increased from 0.4 to 1.6 by using round notches with smaller radii. Katnam et al. [20] followed a similar approach with a toughened adhesive. With smaller notch radii the triaxiality increased but its distribution also became less uniform due to the greater stress concentration around the notch. The experimental work revealed that under conditions of increasing constraint the strength of the adhesive also increased. Cooper et al. [10] observed similar behaviour on a nano- and micro-toughened adhesive using a circumferentially deep notched tensile (CDNT) specimen, with constraint varied by notch depth. Numerical simulations of the tests revealed that H varied between 1 and 1.4, below the level experienced ahead of the crack tip in tapered double cantilever beam fracture tests.

Another triaxial test method, known as the butt-joint or poker chip test consists of two stiff substrates bonded over a given adhesive layer thickness. The stiff substrate restricts the Poisson's contraction of the adhesive layer thereby increasing the stress triaxiality [21]. The failure strain under these conditions is reduced compared with uniaxial tensile tests [21]. Elevated stress triaxialities with the method are limited by premature interfacial failure due to the singularity that exists at the material interface.

In this work it is attempted to experimentally measure the cohesive strength of a nano-toughened adhesive at different levels of stress triaxiality. The procedure combines features of the methods above to achieve values of H that are comparable to adhesive joint fracture, without the stress concentrations seen in other methods. This will enable direct measurement of σ_{max} for accurate modelling of the adhesive layer. Additionally, the measured traction separation curve is examined in detail which allows the conditions for damage initiation and particle decohesion to be identified.

2. Experimental materials and procedures

2.1. Materials

Two materials have been studied in this work. The first is a single part hot cured epoxy based on Epon 828 cured with dicyandiamide. Further components have been added to this formulation to improve its performance but this remains proprietary information. For the remainder of this work, this material shall be referred to as the matrix. The second material is a toughened adhesive, consisting of the same matrix with two grades of core-shell-rubber (CSR) nanoparticles added simultaneously. The first particle is Kaneka MX153 (Kaneka Corporation, USA) with a measured average diameter of 66 nm and the second is Zeon F351 (Zeon Corporation, Japan) with average particle diameters of 200 nm, these particles occupy 16 Vol% and 22 Vol% respectively. TDCB testing on the identical system has shown a 12 fold increase in the fracture energy of the toughened adhesive compared to the neat matrix [22,7,10,23]. A CSR particle consists of a rubber core surrounded by a glassy polymeric shell. The cure schedule for both materials is 180 °C for 90 min.

2.2. Thermal analysis

Differential scanning calorimetry was used to measure the glass transition temperature T_g of the matrix and toughened adhesive. Samples were heated at a rate of 10°/min, and the glass transition was identified by a rapid change in specific heat capacity using the midpoint method [24]. It was found that for the matrix T_g=375.85 K and for the toughened adhesive T_g=380.25 K. Since the values are in relatively close agreement it can be assumed that the mechanical properties of the matrix in the toughened adhesive are equivalent to the bulk matrix [5].

2.3. Bulk deformation behaviour

Uniaxial tension, uniaxial compression and plane strain compression tests were completed on both the matrix and toughened adhesive samples. All tests were conducted at low loading rates and ambient temperature. For each test configuration a minimum of three repeats were performed.

2.3.1. Manufacture of bulk samples

The preparation of bulk samples for each mechanical test method closely followed the guidelines provided by standard BS ISO 15166-2:2000 [25]. Plates of material were cured in aluminium moulds, and then machined to their final geometry. In the case of tensile test specimens the machined surfaces were polished manually using 1200 grit emery paper to remove any defects produced by the machining process.

2.3.2. Uniaxial tension tests

Uniaxial tensile tests were conducted according to ISO 527 with a sample geometry according to the B configuration. The samples were tested in a screw driven tensile testing machine at a cross-head displacement rate of 0.1 mm/min. The load, P, was measured by a 5 kN load cell and the elongation of the gauge length Δl_A and reduction in width Δl_T of the specimen were determined using non-contact video extensometry. This allowed the calculation of axial strain $\epsilon_A = \frac{\Delta l_A}{l_A}$, the transverse strain $\epsilon_T = \frac{\Delta l_T}{l_T}$, Poisson's ratio $\nu = \frac{-\epsilon_T}{\epsilon_A}$, Young's modulus E and yield stress σ_y from each test.

2.3.3. Uniaxial compression tests

The uniaxial compression test setup consisted of two polished platens connected to a mechanical testing machine. A PTFE sheet was placed on each platen and molybdenum disulphide grease was used to lubricate the surfaces. The sample was placed at the centre of the bottom platen and loaded to 50 kN. Samples were produced with a radius and height of 5 mm. The sidewalls of the sample were monitored during compression and it was found that neither buckling nor barrelling occurred. From each test a stress–strain curve was recorded and E and σ_y were calculated.