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<th>Determination of the cohesive strength and toughening mechanisms of a nano-modified adhesive under a triaxial stress state</th>
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Introduction

Our previous work [1] on a nano rubber modified epoxy adhesive suggested that the observed bond thickness effect was due to the level of constraint ($\sigma_{hyd}/\sigma_{eq}$), a measure of the stress triaxiality, in the adhesive layer. In that study tapered double cantilever beam (TDCB) specimens were tested under quasi-static conditions for a range of bond gap thicknesses. The void diameters on the resulting fracture surfaces were measured from which the fracture strain was estimated in each case. The ratio of fracture strains corresponding to different constraint levels was found to agree with the predictions of the Rice and Tracey model.

The current work attempts to further investigate the effects of constraint on adhesive joint fracture. Two experimental test methods are employed (i) the standardised LEFM tapered double cantilever beam (TDCB) test, to measure the mode I fracture energy as function of bond gap thickness and (ii) a recently developed circumferentially deep notched tensile (CDNT) test to determine the cohesive strength and traction-separation behavior at a level of constraint experienced during adhesive joint fracture. Finite Volume simulations of the TDCB and CDNT tests examined the role of the stress triaxiality on the possible failure mechanisms taking place.

Materials and Experimental Procedure

Introduction

Two experimental test methods were employed to examine the adhesive joint fracture behaviour of a nano-toughened epoxy adhesive. All tests were conducted at low loading rates and ambient temperature.

Adhesive

Henkel produced a specially formulated adhesive based on an epoxy matrix. The formulation was toughened by the addition of two populations of core-shell rubber nano-particles. The mean diameters of the particles are 50 nm and 200 nm, which occupy 16 Vol% and 22 Vol% respectively of the total adhesive volume. Standard uniaxial tensile tests were performed on samples machined from a cured sheet of both the toughened adhesive and matrix, according to BS-527:1996 [2]. The mechanical properties obtained and those of the substrate materials used are shown in Table 1.

TDCB tests

TDCB tests were conducted according to the standard [3] to measure the mode I fracture toughness, $G_{IC}$, of the nano-toughened epoxy adhesive as a function of bond thickness. A high yield strength Aluminium alloy, AL 2014 was chosen as the substrate material to prevent plastic deformation of the beams during testing. The surfaces to be bonded were grit-blasted with an alumina grit followed by a chromic acid etch treatment to ensure cohesive failure.

TDCB specimens with bond gap thicknesses ranging from 0.2 mm to 2.5 mm were produced. Side grooves were machined into specimens with a bond gap thickness greater than 0.4mm to ensure mode I fracture. Testing was carried out with a standard tensile testing machine with the load, crack length, and displacement of the crosshead recorded until joint failure.

CDNT Test

The CDNT test method has been developed to measure the maximum cohesive strength and the traction separation behaviour of an adhesive over a range of stress triaxialities similar to those ahead of the crack tip in an adhesive joint. The test involves bonding two cylindrical substrates at a given bond thickness. A notch is then machined circumferentially into the adhesive layer. Appropriate selection of the bond gap thickness and notch geometry produces the desired level of constraint across the remaining adhesive ligament. A typical specimen is shown schematically in Figure 1. Samples with bond gap thicknesses of 1.0mm, 1.6mm and 2.5mm were manufactured with a ligament to bulk ratio of 50%. The specimens were loaded in tension and the displacement and load were recorded. Also a series of tests were completed where the specimens were unloaded prior to failure and the complete load-unload cycle was recorded.

Table 1. Mechanical properties of materials used

<table>
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<tr>
<th></th>
<th>E [GPa]</th>
<th>$\sigma_{yield}$ [Mpa]</th>
<th>UTS [MPa]</th>
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<tbody>
<tr>
<td>Toughened Adhesive</td>
<td>1.76</td>
<td>29.0</td>
<td>45</td>
</tr>
<tr>
<td>Matrix</td>
<td>3.0</td>
<td>63.38</td>
<td>89.7</td>
</tr>
<tr>
<td>AL 2014</td>
<td>72.4</td>
<td>425</td>
<td>475</td>
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Microscopy

During the fracture process, the CSR particles debond from the epoxy matrix, followed by plastic growth of the resultant void. A microscopy study of the TDCB fracture surfaces determined the dilatational void strain, $\varepsilon_f$, prior to fracture for each bond gap thickness. Fractured TDCB specimens were cut to provide sections containing the first 10mm of crack growth. These sections were then gold sputter coated and viewed under SEM. Images were taken along the centreline of the specimen in the direction of crack propagation. The radii of the voids remaining from debonding of the larger ~200nm particles were measured for each bond gap thickness. The average dilatational fracture strain, $\varepsilon_f$ was then computed where $\varepsilon_f = \frac{u_r}{r_0}$, and $u_r$ is the radial displacement of the void and $r_0$ is the initial radius of the nano-particle, i.e. 100 nm in this case.

Experimental Results

Steady state cohesive crack growth occurred for all TDCB tests. The mode I fracture toughness was determined using a corrected beam theory solution given in [3]. It was found that the fracture energy steadily increased from 3400 J/m$^2$ at 0.2mm bond gap thickness up to 5600 J/m$^2$ at 1.6mm beyond which $G_{IC}$ remained relatively constant. The dilatational strain of the void at fracture, $\varepsilon_f$ was compared with $G_{IC}$ for each bond thickness. The extent of void growth prior to failure scaled with fracture energy up to a maximum of 32%, see Figure 2, suggesting that void growth is a significant toughening mechanism.

The traction displacement traces for the three CDNT test geometries can be seen in Figure 3. Each traction separation curve consisted of an initial linear region ended by an abrupt reduction in stiffness, followed by another linear region that continued up to near the peak stress of 52 MPa. From then on displacement increased under a roughly constant stress until failure. The intersection point of the two linear regions is at a higher stress in the loading direction for greater bond gap thicknesses. The series of load-unload tests demonstrated that permanent deformation took place when the specimen was loaded past this intersection point, and that the magnitude of permanent deformation increased when unloaded from a state of greater stress. This intersection point is of interest as it is apparently the initiation of damage in the material under a triaxial stress state.

Numerical Modelling

Numerical simulations of the CDNT and TDCB tests were performed with the OpenFoam Finite Volume software package (version 1.4). A non-linear elastic plastic material model including conventional J2 plasticity governed the adhesive behaviour while all substrates were treated as
being linear elastic. The fracture process was incorporated via a Dugdale cohesive zone model (CZM) specified by the cohesive strength, $\sigma_{\text{max}}$ and the fracture energy, $G_0$. A cohesive strength of 52 MPa, based on the experimental CDNT tests, was selected for all simulations.

The numerical setup reproduced the experimental CDNT traction displacement curve except for the reduction in stiffness noted earlier. The average hydrostatic stress over the ligament area was reported from the numerical model. It was found that the change in slope for all CDNT tests happened at a constant hydrostatic stress of ~24MPa. From microscopy it is clear that during the fracture process significant particle debonding occurs followed by plastic void growth. The reduction in stiffness in the CDNT tests at a constant hydrostatic stress suggests that particle debonding is responsible for the abrupt change in slope. The measured peak stress of 52 MPa corresponds to a stress of 84MPa supported by the matrix with complete debonding. This is based on the area fraction of the matrix to be 62 % since the volume fraction of particles is 38 %. 84 MPa agrees closely with the UTS of the matrix material of 89 MPa. This further suggests the occurrence of particle debonding. H was also calculated prior to the onset of damage and was found to increase from 1.36 to 2.11 with a decrease in bond thickness.

The TDCB tests were simulated incorporating the constant stress Dugdale model. To reproduce the experimental results it was necessary for $G_0$ to be approximately 95% of the experimental fracture energy. The remaining fracture energy was dissipated through a small plastic zone ahead of the crack tip at the free surface of the adhesive layer. Comparison with the experimental load displacement trace and crack length history can be seen in Figure 5. The average constraint immediately ahead of the crack tip was determined to lie between 1.66 and 2.79 for all test configurations. This overlaps the range of stress triaxialities experienced in the CDNT tests.

![Figure 5. Comparison of numerical TDCB tests with experimental](image)

**Conclusions & Future Work**

A specially formulated nano-toughened structural adhesive was developed with Henkel. Under TDCB testing the adhesive demonstrated the classical bond gap thickness dependency with fracture toughness values ranging from 3400 J/m² to 5600 J/m². A CDNT test was developed which found the cohesive strength of the adhesive in terms of constraint. This resulted in the physically realistic definition of a cohesive zone model for use in numerical modelling.

Finite Volume simulations of the CDNT and TDCB tests incorporated a cohesive zone model calibrated from experimental testing. This resulted in an accurate prediction of the experimental data.

Numerical analysis of the CDNT tests indicated that damage in the adhesive layer under triaxial conditions was triggered by a critical hydrostatic stress. With the TDCB simulations the effects of the selected cohesive parameters became clear. As expected [4], the defined ratio of cohesive strength to yield strength of 1.8 prevented a significant diffuse plastic zone from developing before crack advance. This suggests that the mechanisms of material separation including void debonding, void growth and possibly shear banding [5] in the fracture process zone are responsible for the observed fracture toughness.

Extensive microscopy is underway to fully characterise the damage mechanisms taking place throughout the volume of the adhesive layer. With this description the structure property relationship can be fully established by coupling the experimental data with numerical analysis of the adhesive microstructure.

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**References**

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