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The Fracture Behaviour of a Nano-modified Structural Epoxy Adhesive

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1 Introduction

Structural epoxy adhesives typically contain second phase particles to improve their resistance to crack growth. The presence of these particles can dramatically raise the toughness of the system to several times that of the neat epoxy. One of the most common tougheners are core shell rubber (CSR) particles which consist of a glassy shell surrounding a rubbery core. The bulk fracture properties of these systems have been studied by many authors. It is generally accepted that the improvement in toughness is derived from the plastic growth of voids that nucleate from the failure of CSR particles and the development of shear bands between these voids [1]. The development of these mechanisms within the joint are affected by the level of stress triaxiality which depends on many factors including the thickness of the adhesive layer, $h_a$ [2].

The aim of this work is to identify the primary toughening mechanisms that develop during fracture of metal joints that are bonded with these adhesives. This is completed with tapered double cantilever beam (TDCB) fracture tests combined with fracture surface analysis, numerical modelling of the adhesive joints and analytical modelling of the adhesive microstructure. Additionally, a novel test method, the bonded circumferentially deep notched tensile test (CDNT) is used to measure the traction separation behaviour of the adhesive as a function of constraint. The findings of this work also support another study which involves the development of a numerical micromechanical model of the adhesive microstructure. Ultimately, this will evolve into a design tool for the development of improved materials.

2 Experiment

2.1 Materials

Two materials have been used in this work, the first is a single part thermally cured epoxy based resin which shall be referred to as the matrix. The second material is a toughened adhesive, consisting of the same matrix into which two grades of CSR nanoparticles are added. The two CSR grades in this study have average radii, $r_0$, of 33 nm and 100 nm which occupy 16 Vol% and 22 Vol% of the toughened adhesive respectively. The cure schedule for both materials is 180 °C for 90 minutes. The mechanical properties of the matrix and toughened adhesive were determined from standard mechanical tests and the results are shown in Table 1. Experiments on metal joints were only performed on specimens bonded with the toughened adhesive. All tests were conducted at ambient temperature under quasi-static loading conditions.

Table 1: Mechanical properties of the matrix and toughened adhesive

<table>
<thead>
<tr>
<th></th>
<th>$E$ (GPa)</th>
<th>$\nu$</th>
<th>$\mu$</th>
<th>$\sigma_t$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matrix</td>
<td>3.0</td>
<td>0.36</td>
<td>0.28</td>
<td>90</td>
</tr>
<tr>
<td>Toughened</td>
<td>1.79</td>
<td>0.42</td>
<td>0.24</td>
<td>45.2</td>
</tr>
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</table>

2.2 CDNT tests

The CDNT test method has been developed to directly measure the traction separation behaviour of an adhesive at elevated stress triaxialities. 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 provides the desired level of constraint across the adhesive ligament. A schematic of a typical specimen is shown in Fig. 1.

Samples with bond gap thicknesses of 1.0 mm, 1.6 mm and 2.5 mm were manufactured from mild steel substrates. The notch depth was chosen to provide a ligament to bulk area ratio of 50%. The specimens were loaded in
### 2.3 TDCB tests

TDCB tests were conducted according to the standard test method to measure the mode I fracture toughness, \( G_{IC} \), of the toughened epoxy adhesive as a function of bond thickness, \( h_a \). A high yield strength aluminium alloy, Al 2014 was selected as the substrate material. TDCB specimens with \( h_a \) ranging from 0.2 mm to 2.5 mm were produced. Side grooves were machined into the adhesive layer of specimens with a bond gap thickness greater than 0.4 mm to ensure mode I fracture. Testing was completed on a standard tensile testing machine. The load, crack length, and displacement of the crosshead were monitored until joint failure.

### 2.4 Microscopy

Sections from the TDCB and CDNT specimens were viewed under scanning electron microscopy (SEM). The fracture surfaces of specimens from both the CDNT and TDCB tests were similar, consisting of a series of voids nucleated by the failure of the CSR nano-particles. The average radius of the voids, \( r_v \), on the fracture surface of the TDCB specimens was determined for each bond gap thickness. The extent of void growth was expressed in terms of the fracture strain, \( \varepsilon_f \), which is calculated as \( \varepsilon_f = (r_v - r_0)/r_0 \).

### 2.5 Results

Three representative curves of the CDNT tests are shown in Fig. 2. For all tests a peak strength of near 52 MPa was recorded. There is also an abrupt kink in the traction separation curve for each test. The load-unload tests indicated that the kink is the initiation of damage in the material since permanent deformation begins at this point. The TDCB tests followed the classical bond gap thickness dependency, whereby, the fracture energy increased steadily with bond gap thickness from 3400 J/m\(^2\) to a maximum of 5600 J/m\(^2\) after which the fracture energy reduced slightly, as shown in Fig. 3. The extent of void growth in terms of the fracture strain, \( \varepsilon_f \), for each particle family is shown in Fig. 3. Interestingly, the extent of plastic void growth for both particle families, described by \( \varepsilon_f \), scales with the fracture energy of the joint, indicating that plastic void growth is a significant toughening mechanism for this adhesive.

### 3 Numerical Modelling

Finite Volume numerical simulations of the CDNT and TDCB tests were performed with the OpenFoam software package (version 1.4). A non-linear elastic plastic material model including conventional J2 plasticity governed...
the adhesive behaviour while all substrates were modelled as 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. This number was calculated based on the experimental stress-strain curve.

The stress triaxiality was defined in terms of a constraint parameter, $H$, where $H = \sigma_{\text{hyd}}/\sigma_{\text{eq}}$. The average value of constraint, $H_{\text{avg}}$, was calculated in the TDCB and CDNT tests. For both methods, $H_{\text{avg}}$ is greater at lower bond gaps: for the TDCB tests $H_{\text{avg}}$ increased from 1.64 to 2.79 and for the CDNT tests $H_{\text{avg}}$ was between 1.37 and 2.09. The range of $H_{\text{avg}}$ in both methods is comparable, thus it is assumed that the failure mechanisms in each test develop in a similar manner which allows relation of results between the TDCB and CDNT tests.

The behaviour of the kink reported from the CDNT tests, was analysed in the numerical simulations. From experiments it was found that with increasing constraint (lower $h_b$) the kink occurred at a lower applied stress. However, the numerical simulations revealed that the kink occurred at constant hydrostatic stress of 23 MPa across the range of specimens. This strongly suggests that the CSR particles are failing at this point since both debonding and cavitation are controlled by a critical hydrostatic stress.

Detailed analysis of the CDNT simulations indicated that no plasticity occurred prior to failure of the CSR particles. In addition, simulation of the TDCB tests revealed that nearly all of the fracture energy was dissipated within the fracture process zone, and not by diffuse von Mises plasticity in the adhesive layer. This is a consequence of the prescribed ratio of $\sigma_{\text{max}}/\sigma_y = 1.79$ which prohibits the development of a classical plastic zone around the crack tip.

4 Process Zone Modelling

The quantity of energy dissipated in the fracture process zone can be approximated using the model developed by Huang and Kinloch [3, 4]. The model is employed in this work to examine the source of the observed toughness. Huang and Kinloch proposed that the fracture energy of a particle filled epoxy, $G_{cc}$, is the sum of three distinct contributions: the fracture energy of the matrix $G_{cm}$; the energy dissipated in shear bands, $\Delta G_s$; and the energy dissipated by plastic void growth, $\Delta G_v$, such that

$$G_{cc} = G_{cm} + \Delta G_s + \Delta G_v.$$  \hfill (1)

The contribution from void growth is given by

$$\Delta G_v = 2 r_y \sigma_{\text{hyd}} (V_v - V_p)$$  \hfill (2)

and from shear bands as

$$\Delta G_s = 0.5 r_y [4 \pi / 3 V_p]^{1/3} - 54/35 V_p \sigma_y \gamma_f$$  \hfill (3)

where $\gamma_f$ denotes the shear failure strain taken to be 0.75 from [5], $V_p$ is the volume fraction of particles and $V_v$ is the volume fraction of voids. The radius of the process zone, $r_y$, is calculated according to

$$r_y = \left(1 + \mu / \sqrt{3}\right)^2 K_{ym}^2 r_{ym}$$  \hfill (4)

where $\mu$ accounts for the sensitivity of matrix yielding to hydrostatic stress, $K_{ym}$ is a stress concentration factor that depends on $V_p$ which is given as 3.5 in [3] and $r_{ym}$ is the Irwin plastic zone size given by

$$r_{ym} = \frac{1}{6 \pi (1 - \nu_m^2)} \sigma_y^2$$  \hfill (5)

where $E_m$, $\nu_m$ and $\sigma_y$ are the Young’s modulus, Poisson’s ratio and tensile yield stress of the matrix respectively. Liang and Pearson [6] suggested that ($V_v - V_p$) should be calculated by:

$$(V_v - V_p) = \frac{V_v}{V_v + V_m} - \frac{V_p}{V_p + V_m}$$  \hfill (6)

where $v$ are $V$ and particle volume.

The average void volume is determined from the measured void diameters on the fracture surface, assuming spherical void growth and the average matrix volume $V_m$ is expressed as

$$v_m = \frac{V_p (1 - V_p)}{V_p}.$$  \hfill (7)

In the original model, the hydrostatic stress during plastic void growth is calculated as $\sigma_{\text{hyd}} = 0.5 \sigma_y$. After particle failure the matrix alone supports the applied load. Thus, in this case the mechanical behaviour can be described as a porous elasto-plastic solid, with a porosity equivalent to the volume fraction of particles. Jeong [7] developed the following yield function for porous materials that consist of pressure sensitive matrices

$$\Phi = \left[\frac{\sigma_m}{\sigma_y}\right]^2 + \zeta^2 \left[2.7 V_p \cosh \left(0.95 \frac{3 + 2\sqrt{3} \mu}{2\sqrt{3} \mu} \log \zeta \right) \right]$$

$$- \zeta^2 (1 + 35 V_p^2)$$

$$= 0$$  \hfill (8)

where

$$\zeta = \left(1 - \sqrt{3} \mu \frac{\sigma_{\text{hyd}}}{\sigma_y} \right).$$  \hfill (9)

To satisfy Eq. (8) when $H$ is between 1.4 and 2.8 requires that $\sigma_{\text{hyd}}$ is between 23 MPa and 25 MPa during void growth. We propose that these values for hydrostatic stress offer a more accurate description of the process. Three scenarios were considered for predicting $G_{cc}$ from the model:
1. \( G_{cc} = G_{cm} + \Delta G_v \) with \( \sigma_{hyd} = 0.5 \sigma_v = 45 \) MPa
2. \( G_{cc} = G_{cm} + \Delta G_v \) with \( \sigma_{hyd} = 25 \) MPa and
3. \( G_{cc} = G_{cm} + \Delta G_v + \Delta G_s \) with \( \sigma_{hyd} = 25 \) MPa.

Calculation of Eq.(4) results in \( r_{cc} = 0.373 \) mm, since the process zone cannot grow into the substrates \( r_{cc} \) is taken to be 0.1 mm and 0.2 mm when \( h_a = 0.2 \) mm and \( h_a = 0.4 \) mm respectively.

### 4.1 Results

The results obtained from the model according to each scenario are shown in Fig. 4. By comparing scenario 1 and 2 it is clear that \( \sigma_{hyd} = 25 \) MPa instead of \( \sigma_{hyd} = 45 \) MPa results in a substantially lower value for \( \Delta G_v \) that agrees more closely with experiments.

For \( h_a \leq 0.4 \) mm the scenario 2 prediction is significantly below the measured fracture energy. For these lower bond gap thicknesses \( \Delta G_v \) is a robust upper bound, therefore partnering mechanisms, such as shear bands must be taking place. For \( h_a > 0.4 \) mm, scenario 2 agrees closely with the experimental data, however the inclusion of \( \Delta G_s \), as shown in scenario 3 severely overestimates the experiments. Since there is convincing experimental evidence for the void growth mechanism we conclude that the energy dissipated by shear bands is not significant at these greater bond gap thicknesses, but it must play a significant role at lower thicknesses. Conclusive support for these findings requires a detailed microscopy study through the thickness of the adhesive layer.

**Figure 4: Fracture energy predictions with the Huang and Kinloch model**

### 5 Conclusions

The bonded CDNT test has been presented as a useful method to determine the mechanical behaviour of structural adhesives under conditions of high stress triaxiality. Based on the experimental results the cohesive triaxiality, \( \sigma_{max} \) and initiation of damage within the adhesive layer has been identified under conditions that are similar to those at the crack tip. This facilitated the development of a realistic numerical model of fracture defined by measured parameters. The model demonstrates that the most of the fracture must be dissipated in the process zone. Modelling of the process zone with the Huang and Kinloch model suggests that at lower bond gap thicknesses both shear bands and void growth contribute to the fracture energy. However, as the bond gap thickness increases void growth becomes the dominant toughening mechanism.

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**References**


