<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>The Bond Gap Thickness Effect on the Fracture Toughness of Nano-Toughened Structural Epoxy Adhesives</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Authors(s)</strong></td>
<td>Cooper, V.; Ivankovic, Alojz; McAuliffe, David; et al.</td>
</tr>
<tr>
<td><strong>Publication date</strong></td>
<td>2010-02-19</td>
</tr>
<tr>
<td><strong>Conference details</strong></td>
<td>33rd Annual Meeting of the Adhesion Society, Daytona Beach, Florida, February 21-24, 2010</td>
</tr>
<tr>
<td><strong>Publisher</strong></td>
<td>Adhesion Society</td>
</tr>
<tr>
<td><strong>Item record/more information</strong></td>
<td><a href="http://hdl.handle.net/10197/4763">http://hdl.handle.net/10197/4763</a></td>
</tr>
</tbody>
</table>
THE BOND GAP THICKNESS EFFECT ON THE FRACTURE TOUGHNESS OF NANO-TOUGHENED STRUCTURAL EPOXY ADHESIVES

V. Cooper, A. Ivankovic, A. Karac, D. McAuliffe, N. Murphy, Z. Tukovic.
School of Electrical, Electronic and Mechanical Engineering,
University College Dublin, Ireland

Introduction
This work employs combined experimental and numerical studies to investigate the effect of bond gap thickness on the fracture behaviour of a nano-toughened epoxy adhesive produced by Henkel. Tapered-double cantilever-beam (TDCB) joints were subjected to a constant low loading rate. The mode I fracture behaviour of the joints was investigated as a function of bond gap thickness, which was varied from 0.25 to 2.5 mm. A detailed analysis of the fracture surfaces was carried out using scanning electron microscopy (SEM) and variations of the microstructural features with the bond gap thickness, and corresponding constraint, were revealed. The Rice and Tracey void growth model is used to relate the local plastic strain calculated by measuring the size of the voids on the fracture surfaces with the constraint calculated numerically for each bond gap thickness. It was shown that the difference in constraint imposed by different bond gap thicknesses is responsible for the observed dependency of $G_{IC}$.

Background
Many studies have been carried out to investigate the effect of bond gap thickness on the fracture behaviour of adhesive joints [1-6]. In most of these studies the fracture energy of the adhesive joints was reported to reach a maximum value, $G_{IC}$, at some bond gap thickness $h_{am}$. Kinloch and Shaw [3] concluded that $G_{IC}$ depends on the size of the plastic zone, and that the constraining effects from the two adherents control the plastic zone size. When the bond thickness is less than $h_{am}$ the plastic zone can not fully develop due to the restriction imposed by the substrates. When the bond thickness becomes larger than $h_{am}$ the plastic zone is no longer constrained by the substrates and its size remains independent of the bond thickness. The maximum, $G_{ICmax}$, is therefore reached when the adhesive layer thickness and the plastic zone diameter, $2r_p$, are approximately equal. Also, $G_{ICmax}$ should equal the mode I fracture toughness of the bulk adhesive specimen, $G_{IC(bulk)}$. Thus, the adhesive thickness, $h_{am}$ can be expressed as [7]:

$$h_{am} = 2r_p = \frac{1}{\pi} \frac{E G_{IC(bulk)}}{\sigma_y}$$

(1)

where, $r_p$ is Irwin’s first order estimate of the plastic zone size (plane stress), $E$ is the adhesive modulus, and $\sigma_y$ is the uniaxial yield strength of the adhesive.

However, the applicability of the classical plastic zone size argument is found to be questionable. The TDCB test is not a $K_I$ dominant test due to the presence of the adherents and hence the prediction based on equation 1 may not be accurate. Also, this approach implies that total fracture energy is the sum of a local intrinsic contribution ($G_{IC}$) coming from the fracture process zone (FPZ) and a non-local extrinsic contribution from the crack tip plastic zone ($G_p$). The value of $G_p$ should be measured at the bond thickness equal to the size of the FPZ and hence is difficult to define, while $G_p$ increases with increasing bond thickness from zero value at FPZ to its maximum at $h_{am}$. In the present work, numerical simulations were employed with a cohesive zone model (CZM) to analyse the fracture of TDCB specimens. It was found that the size of the plastic zone was negligible due to the low value of cohesive strength, which was required to accurately reproduce experimental results. Hence, the plastic zone argument could not be used to explain the bond gap effects on the fracture toughness. It should be noted that $K_I$ dominant conditions are further eliminated by the presence of the CZM. In the present work it was shown that fracture toughness cannot be partitioned and the bond gap thickness effects were attributed to the effects of the constraint on the plastic void growth.

Materials and Experimental Procedures

Adhesive and substrate materials
The adhesive used is a single part, hot cured structural epoxy adhesive (Henkel Lab. ref. 3019-98) containing two core shell particle grades (~50nm and 200nm diameter) and a liquid co-toughener. Two substrate materials are employed: (i) Aluminium 2014 - a high yield strength alloy and (ii) Steel EN24T - a high quality, high strength, alloy steel. The basic mechanical properties of the adhesive and the substrates are given in the Table 1.

<table>
<thead>
<tr>
<th>Material</th>
<th>E [GPa]</th>
<th>UTS [MPa]</th>
<th>$\sigma_{pul}$ [MPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al 2014</td>
<td>72.4</td>
<td>472</td>
<td>425</td>
</tr>
<tr>
<td>Steel EN24T</td>
<td>205</td>
<td>875</td>
<td>1000</td>
</tr>
<tr>
<td>Henkel 3019-98</td>
<td>1.46</td>
<td>38.5</td>
<td>28</td>
</tr>
</tbody>
</table>

TDCB tests
In the current work, the mode I fracture behaviour of the adhesive joints was examined using low rate TDCB tests with various bond gap thicknesses ranging from 0.25 mm to 2.5 mm. Two TDCB setups were used: (i) Steel substrates of 25.4 mm width [8] and (ii) 10 mm wide aluminium substrates [9]. Steel substrate TDCB tests were conducted at 0.3 mm/min loading rate, while the aluminium substrate TDCB specimens were subjected to 0.5 mm/min rate. The exact geometries are illustrated in Figure 1. The tests were carried out at room temperature and records of the load and the displacement at different increments of the crack length were obtained. At least three repeats for each
joint system were tested.

**Experimental Results**

Steady state crack growth behaviour was observed in all tests. $G_{IC}$ was calculated using the corrected beam theory approach given in [9]. For tests with a bond thickness less than 1mm, mode I cohesive failure was observed. However when the bond gap thickness was increased above 1mm, a slanted fracture gradually developed along the specimen. This was eliminated through the introduction of side grooves to the adhesive layer. It is interesting to note that the values of $G_{IC}$ were in very close agreement for specimens both with and without side grooves. An increase in $G_{IC}$ with bond gap thickness was observed and is plotted in Figure 2. Both configurations gave similar values of $G_{IC}$ for a given bond gap thickness. The average value of $G_{IC}$ was found to increase from 2600 J/m² at 0.25 mm bond gap thickness to 5800 J/m² at 1.3 mm, and remain constant for larger bond gap thicknesses.

![Figure 1: Illustration of the TDCB geometries: (a) BS 7991:2001 (m = 2mm) and (b) ASTM D3433:1999 (m = 3.543mm). All dimensions are in mm.](image)

**FV Modelling**

Numerical modelling of the two TDCB test setups was performed using the FV method (OpenFOAM - version 1.4 [10]). Both the substrate and the adhesive were modelled using the conventional J2 elastic-plastic material model. Only the 10mm wide BS results are presented here. The CZM was defined using a two-parameter Dugdale model, with parameters being the adhesive fracture energy $G_{IC}$ and the maximum cohesive stress, $\sigma_{max}$. The bond gap effect on fracture toughness in the TDCB samples was reproduced numerically only when a value of $G_{IC}$ at a given bond gap thickness was taken equal to the experimentally obtained value given in Figure 2. The comparison between measured and predicted load and crack length histories are shown in Figure 3. Numerical simulations of TDCB specimens showed a strong sensitivity to the value of $G_{IC}$ while $\sigma_{max}$ had less pronounced effects. A sensitivity analysis was carried out to investigate the effect of cohesive strength and it was found that the results were less sensitive to variations in $\sigma_{max}$ for smaller bond gap thicknesses (i.e. higher constraint).

![Figure 3: Comparison of numerical and experimental BS TDCB results. $G_{IC} = \text{experimental, } \sigma_{max} = 38.5\text{MPa (UTS).}$](image)

In the current work, the constraint factor $H$ is used as a measure of stress triaxiality. It is defined as the ratio of the hydrostatic stress, $\sigma_{hyd}$, to the von Mises equivalent stress, $\sigma_{eq}$. A number of ways, all giving reasonably close results, of averaging $H$ for the TDCB specimen were investigated [11]. In the results shown below, an average value of $H$ is taken over the crack front line just prior to crack initiation. In Figure 4, both $G_{IC}$ and $H$ are plotted against bond gap thickness. It can be seen that the point at which $G_{IC}$ begins to plateau corresponds to the point at which $H$ also begins to level off. The vertical line corresponds to the point at which $G_{IC}$ becomes independent of $H$ and also where $G_{IC}$ becomes independent of the bond gap thickness. Hence, a $G_{IC}$ dependency on $H$ is demonstrated.

**Microscopy**

The toughening of the current adhesive is achieved mainly through the addition of core shell nano-particles. The main toughening mechanism results from the debonding of these particles from the epoxy matrix followed by plastic void growth. According to the Rice and Tracey model with a
constant critical void radius, the ratio of two different fracture strains, $\varepsilon_f$ and $\varepsilon_f^i$, at different constraints, $H_f$ and $H_f^i$, is given as:

$$\frac{\varepsilon_f}{\varepsilon_f^i} = \exp[0.5(H_f - H_f^i)] \quad (2)$$

The strain can be approximated as $\varepsilon_f = u_r/r_0$ where $u_r$ is the final radial displacement of the void and $r_0$ is the original radius of the void (i.e. the particle size). A number of large voids were selected from the images given in Figure 5, and the diameters measured.

![Figure 4: Plot of GIC and H against bond gap thickness.](image)

The reason for this is that the large voids correspond to voids that were ‘cut’ by the crack through the middle. Moreover, since the adhesive has two families of particles, it is more appropriate not to average but to choose voids of similar sizes. This gave an average void radius of 112nm for the 0.4mm sample and 135nm for the 2.0mm sample. For a given $r_0$ value of 100 nm, the ratio of $\varepsilon_f \varepsilon_f^i \varepsilon_f^i = 0.34$.

The constraint factors for the two bond gap thicknesses are calculated as $H_{0.4} = 2.2$ and $H_{2.0} = 1.53$, giving the value of the RHS of equation 2 equal to 0.366. These results confirm that the Rice and Tracey model is applicable. The large differences in the plastic strains for the two bond gap thicknesses (factor of almost 3) as caused by the difference in constraint can be used to explain the differences in the corresponding fracture toughness since at a given bond thickness $G_{IC} \propto \sigma_f e_\delta F P Z$, where $\sigma_f$ is the fracture stress, $e_f$ the fracture strain and $\delta F P Z$ is the size of the FPZ.

![Figure 5: Micrograph (x10k) taken from the 0.4mm (left) and 2.0mm (right) bond gap thickness specimens.](image)

Conclusions

This work has offered a valuable insight into the effects of crack-tip constraint on the fracture toughness. It was demonstrated that $G_{IC}$ cannot be partitioned and the bond gap thickness effects were attributed to the effects of the constraint on the plastic void growth. The applicability of the classical plastic zone size argument was found to be questionable. Instead, it was shown that most of the deformation (and energy dissipation) occurs locally in a small region ahead of the crack tip known as the fracture process zone. The Rice and Tracey void growth model was used to analyse the size of the voids on the fracture surfaces and it was shown that the relatively small change in constraint, $H (2.3 – 1.53)$, can result in a large increase in $G_{IC}$ (2.8 – 6 kJ/m²).

It is believed that a micromechanical analysis (e.g. Gurson-Tvergaard-Needleman (GTN) or a unit-cell approach) will help to give a better understanding of the fracture behaviour of nano-toughened structural adhesives.

Acknowledgments

The authors wish to acknowledge the funding and support received from Henkel Loctite Ireland and The Irish Research Council for Science Engineering and Technology.

References

10. www.OpenFoam.com