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## Test Method

# Improvement of adhesive toughness measurement

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## ABSTRACT

The double cantilever beam (DCB) method for adhesive toughness measurement was improved by incorporating a sufficiently sharp crack made by a wedge-tapping method. A known route to producing cracks via loading–unloading cycles was proved unreliable because the cycles produced plastic deformation in the adhesive where new cracks propagated. Abnormally high toughness values with large standard deviations were obtained with cracks made by embedding a non-sticky insert. Only instantly propagated cracks made by tapping were sufficiently sharp to produce reproducible, accurate toughness measurements. However, toughened resin was insensitive to crack sharpness. A crack length to adherend length ratio of 0.2–0.5 is recommended.

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## 1. Introduction

Because catastrophic failure often starts from a micro-crack, fracture toughness - the resistance of a material to the propagation of a sufficiently sharp crack - is a critical material property for brittle resins. Thermosetting resins such as epoxy are widely used adhesives to join metallic adherends. The adherends have different thermal expansion coefficient to the polymeric adhesives, causing micro-cracks within the adhesive or between adhesive and adherends in use due to weathering [1]. These microcracks are highly sensitive to stress concentration encountered in service, leading to catastrophic failures of adhesive joints.

The adhesive toughness of bonded joints can be evaluated in mode I, mode II or mode III as shown in Fig. 1. Of these, mode I is most commonly used in practice because (i) it is the cause of major catastrophic failures [1], and (ii) the sample preparation and testing cost of mode I is the lowest.

ISO and ASTM provide two testing geometries for Mode I: double cantilever beam method (DCB) and tapered double cantilever beam method (TDCB) [2,3], in both of which it is advised to make a precrack by embedding a thin film in the adhesive. Figs. 2 and 3 show the geometries of DCB and TDCB, respectively. Since the manufacturing cost of DCB is substantially lower than TDCB, DCB is more popular in practice [3–8]. Although the details of producing a sharp crack are not always available [9], there are studies which demonstrated the creation of cracks by a loading–unloading method [10–12]. In a typical test, a pair of bonded adherends experienced a number of loading–unloading cycles, each of which propagated a new crack. An adhesive toughness value was calculated for each cycle using the critical load, the ratio of crack length to the adherend length, the sample width, the adherend modulus, etc.

Previous research shows that the presence of a sufficiently sharp crack is a prerequisite for measuring the fracture toughness of brittle resins [9]. Hypotheses made in the current study include (a) a sufficiently sharp crack is essential to the accuracy and reproducibility of the adhesive toughness testing and (b) an appropriate ratio of crack length to adherend length might be necessary to obtain reasonable standard deviations. We will improve the DCB method to obtain

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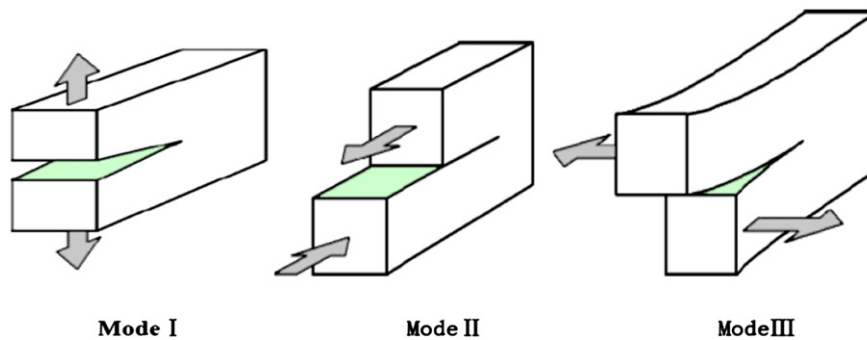


Fig. 1. Schematic of failure modes.

accurate, reproducible results by examining the loading–unloading method, studying how to produce a sufficiently sharp crack and what is the effect of crack sharpness on the test results, and investigating whether there is an effective range for the ratio of crack to adherend length.

## 2. Experiment

### 2.1. Material and accessories

Epoxy resin diglycidyl ether of bisphenol A (DGEBA, EEW 182–196 g/equiv, Araldite-F) was supplied by Ciba–Geigy, Australia. Liquid-rubber, amine terminated butadiene-acrylonitrile copolymer (Hycar 1300 × 35), was provided by Noveon Inc. Silicon grease and sodium hydroxide were purchased from Sigma–Aldrich Co. Jeffamine D230 (denoted as J230) was provided by Huntsman Singapore. The spherical silica nanoparticles (Nanopox F400) were supplied as a colloidal sol (40 wt%) in epoxy by Hanse Chemie AG, Germany.

Adherends were manufactured by a local workshop from aluminum 6060 which has Young's modulus 69 GPa and yield strength 187 MPa. The adherend with dimensions of 150 × 25 × 10 mm were fabricated according to ASTM 3433-99[3] (there was no ISO standard when this work was started). 0.1-mm thick copper shim was used to control the adhesive thickness. 40- $\mu$ m thick, non-sticky paper was purchased locally.

### 2.2. Substrate and adhesive preparation

The thickness and width of each adherend were measured before bonding using a micrometer and a vernier caliper. The substrate was rubbed to remove metal oxide and

contamination using an electric sander. Acetone and cotton balls were used to clean and remove particles produced by polishing. The substrates were then immersed in a 20 wt% sodium hydroxide solution for 15 min. The substrates were removed from solution and cleaned again using acetone and distilled water.

Brass shim and the side surfaces of adherends were coated with a thin layer of silicone grease. Two brass shims were placed at the ends of the adherends to control the adhesive thickness. Epoxy was mixed with hardener J230 using a magnetic stirrer. The mixture was applied on the top surface of the adherends, followed by degassing to remove bubbles. One adherend was carefully turned over and placed on the right top of the other. If required, a non-sticky paper was placed between the adherends to produce a precrack (see Section 2.3.2). The curing process included holding at 80 °C for 30 min, taking 60 min increasing to 100 °C and 90 min to 120 °C, and keeping at 120 °C for 16 h.

### 2.3. Creation of cracks

Since the crack sharpness poses a significant effect on the toughness of brittle resins [9], this study produced three types of cracks by the following methods to identify the effect of crack sharpness on the adhesive toughness.

#### 2.3.1. Crack produced by loading–unloading cycles

A non-sticky paper was embedded in an adhesive joint as a precrack. Tensile loading was applied at 0.5 mm/min perpendicularly to the bonding plane until a crack propagated, which was then unloaded by winding the upper jaw back to the starting level; this cycle was repeated a few times. Multiple toughness values were collected from one sample using these cycles.

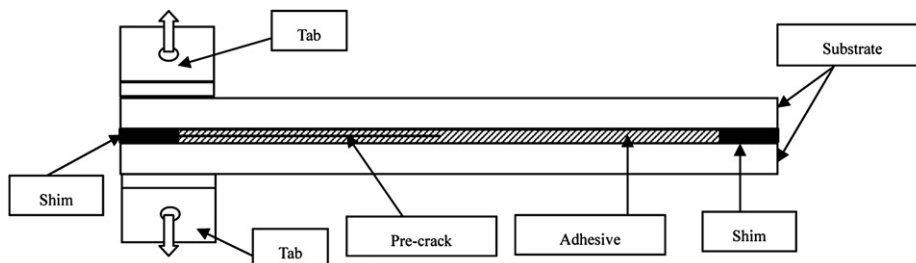


Fig. 2. Double cantilever beam specimen with aluminum substrates.

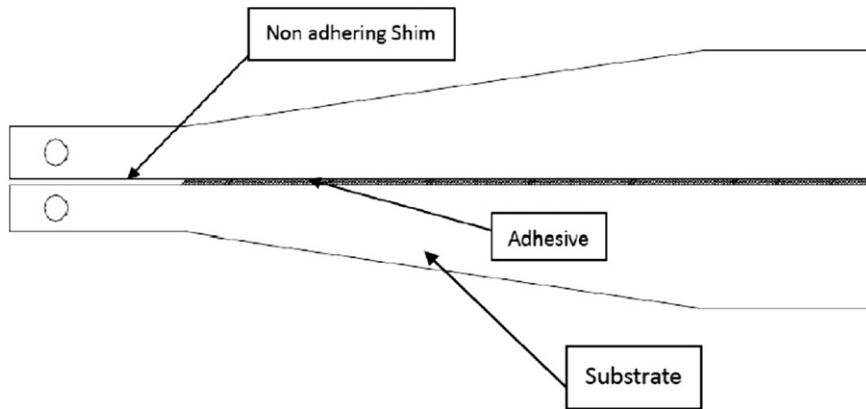


Fig. 3. Tapered double cantilever beam (TDCB) Specimens.

### 2.3.2. Crack made by embedding non-sticky film

Crack made by embedding non-sticky film - a well-known method [2,3,6,7]. As shown in the bottom part of Fig. 4, a non-sticky paper of 40- $\mu\text{m}$  in thickness was placed between the adherends during fabrication as described in 2.2 to create a precrack. Obviously, the film thickness affects the crack sharpness.

### 2.3.3. Crack made by wedge-tapping

In three-point-bending testing, reproducible, accurate fracture toughness values were obtained with the presence of a sufficiently sharp crack made by tapping a razor blade into an epoxy resin [9]. Hence, in this study we produced a sharp crack in the adhesive by a wedge-tapping method. After a desired length of precrack was made by embedding a non-sticky paper, brass shims were removed from the joint. While a third of the DCB length was firmly held from the bottom by a bench vice, a wedge was inserted and tapped to produce an instantly propagated crack as shown in the top image of Fig. 4. Specifically, a layer of plaster coating was put on each side of the joint, and a wedge was placed on the top of the sample and tapped using a hammer until a crack propagated visibly to the desired length. The propagated crack showed a crescent-like shape and was slightly longer than the embedded crack. The tapped crack length could be roughly controlled by adjusting the tapping force.

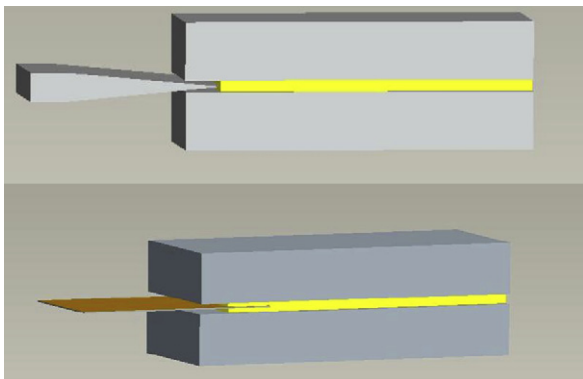


Fig. 4. Pre-crack by wedge-tapping and non-sticky paper.

The sample was then mounted on an Instron machine for measurement. After the sample was fractured, the crack tip points were marked as in Fig. 5 to measure crack length using a digital vernier caliper.

### 2.4. Calculation

The  $G_{Ic}$  values of DCB specimens with load-blocks were calculated as below according to ISO25217 'corrected beam theory' which is considered an improvement over ASTM D3433 [3].

$$G_{Ic} = \frac{3P\delta}{2B(a + |\Delta|)} \cdot \frac{F}{N}$$

$$F = 1 - \frac{3}{10} \left( \frac{\delta}{a} \right)^2 - \frac{3}{2} \left( \frac{l_1 \delta}{a^2} \right)$$

$$N = 1 - \left( \frac{l_2}{a} \right)^3 - \frac{9}{8} \left[ 1 - \left( \frac{l_2}{a} \right)^2 \right] \frac{l_1 \delta}{a^2} - \frac{9}{35} \left( \frac{\delta}{a} \right)^2$$

where:

$\alpha$  = Crack length, mm

$P$  = load measured by the load-cell of the Instron machine, N

$\delta$  = displacement of the cross-head of the test machine, mm

$B$  = Specimen Width, mm

$\Delta$  = crack-length correction, mm. Since only an instantly propagated crack is used,  $\Delta$  is omitted.

$F$  = large-displacement correction

$N$  = load-block correction

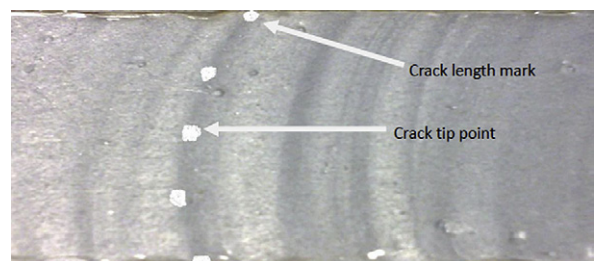


Fig. 5. Crack length marks and crack tip points of a fractured adherend.

$l^1$  = distance from the centre of the loading pin to the loading-block which is a tab in Fig. 2, mm  
 $l_2$  = distance from the pin centre to the loading-block edge, mm

### 3. Results and discussion

#### 3.1. Loading–unloading testing

Since a number of studies obtained fracture toughness by loading–unloading DCB specimens [10–12], we examined the feasibility of this method for testing the adhesive toughness of neat epoxy. Applying four cycles of loading–unloading to a pair of aluminum adherends created four peaks of load shown in Fig. 6; each peak corresponds to a crack. Crack 1 was made by embedding a non-sticky paper. The 1st loading which propagated Crack 1 produced plastic deformation to the adhesive in front of Crack 1; the Crack 1 developed into Crack 2 through the 1st unloading; that is, there is plastic deformation around Crack 2. The other cracks were produced and propagated during the subsequent loading–unloading cycles. In Fig. 6, load increases linearly with displacement for Cracks 1 and 2, corresponding to brittle fracture, while loads for Cracks 3 and 4 show a plateau which implies that substantial plastic deformation occurred. The toughness values calculated from the four cycles are found to obviously reduce with increase in crack length, as shown in Fig. 7; a similar trend was observed for repeated experiments. The last three cracks created by loading–unloading are supposed to be similarly sharp and should produce similar toughness values, contradictory to the experimental data in Fig. 7. This is explained in light of plastic deformation of the adhesive caused by the loading–unloading cycles. Tensile load is applied through the crosshead and grips to the DCB at 0.5 mm/min. Since the Young's modulus of aluminum, 69 GPa, is substantially higher than that of epoxy, 2–3 GPa [13,14], the adhesive undergoes much more deformation than the adherends during the cycles. Hence, a loading that had propagated a crack produced plastic deformation to the subsequently propagated cracks, which is schematically illustrated in Fig. 8. Under loading, Crack 1 was propagated

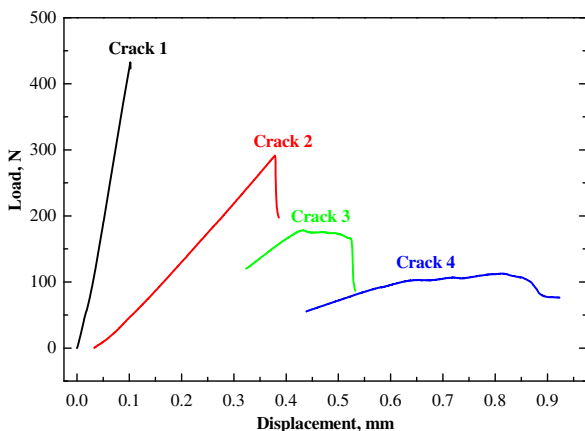


Fig. 6. Load-displacement curve of loading–unloading cycles.

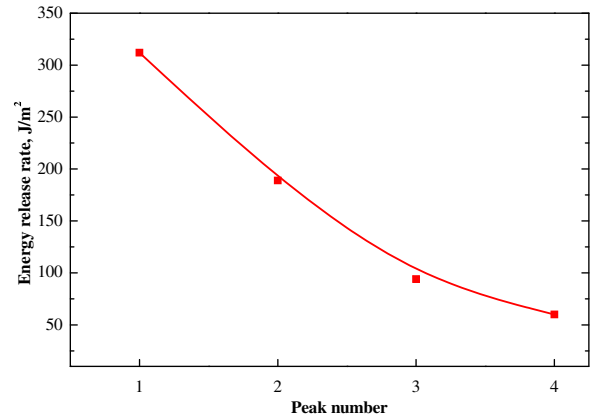


Fig. 7. Adhesive toughness of neat epoxy tested by loading–unloading cycles.

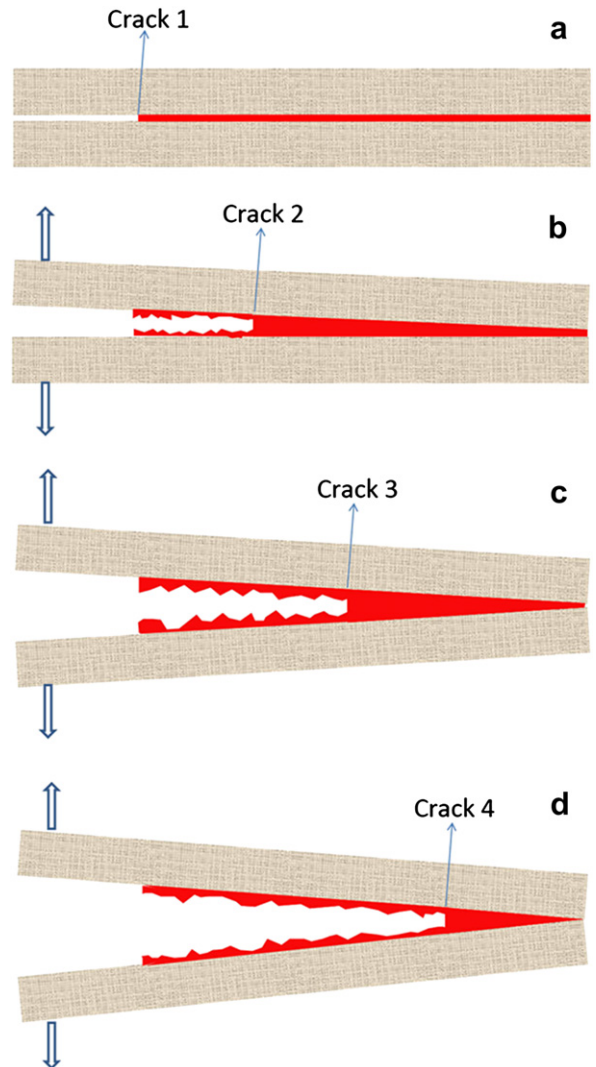


Fig. 8. Schematic of the adhesive plastic deformation during loading–unloading cycles, assuming no elastic deformation of adherends.

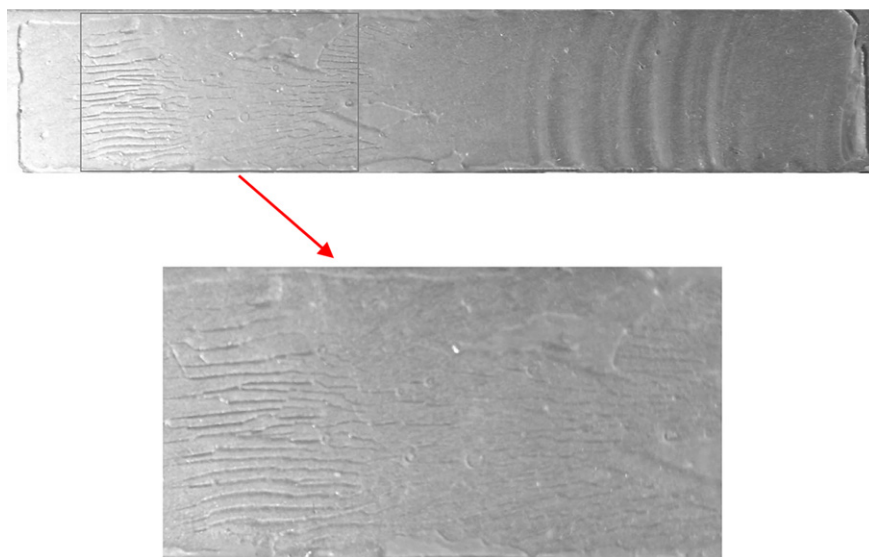


Fig. 9. Fracture surface of adherend showing plastic deformation caused by loading–unloading cycles (rectangle in the top photo is magnified in the bottom one; crack propagates from right to left).

to produce Crack 2 in Fig. 8b followed by unloading; then it was loaded again to produce Crack 3 in Fig. 8c followed by unloading; Crack 4 was similarly made. Before being loaded for propagating into Crack 3, Crack 2 had undergone plastic deformation caused by the propagation of Crack 1. Similarly, Crack 3 and 4 had both undergone plastic deformation before being loaded for propagation. The later a crack propagated, the more plastic deformation it experienced. Thus, Peak 4 shows more plateau than Peak 3 in Fig. 6. This is confirmed in Fig. 9 where the Crack 4 zone is magnified showing obvious plastic deformation, while similar deformation was not found for Crack 1 and 2. Once plastic deformation occurs before testing, the adhesive toughness measured will be inaccurate.

Since ISO25217 recommends a crack made by the 1st loading–unloading cycle for testing [3], five pairs of neat epoxy joints were tested with cracks propagated by one loading–unloading cycle. The measured adhesive toughness  $241.2 \pm 12.6 \text{ J/m}^2$  is higher than the following values obtained from embedded cracks and propagated cracks; which infers that the cracks produced by loading–unloading cycles are insufficiently sharp. Since polymeric adhesive is viscoelastic, the sharpness of a crack depends on its propagation rate - a crack made by loading–unloading at 0.5 mm/min is certainly blunter than an instantly propagated crack made by tapping.

### 3.2. Effect of crack sharpness on toughness of neat epoxy and its toughened resin

In previous research, an instantly propagated crack made by tapping was proven sufficiently sharp for reproducible, accurate fracture toughness measurement on neat epoxy and its toughened resins [9]. Since a crack made by embedding a non-sticky paper is a standard recommended procedure, we compared the effect of this crack with an

instantly propagated crack on the adhesive toughness of neat epoxy and its toughened resins.

Fourteen pairs of neat epoxy-bonded adherends were prepared with a varying ratio of crack to adherend length

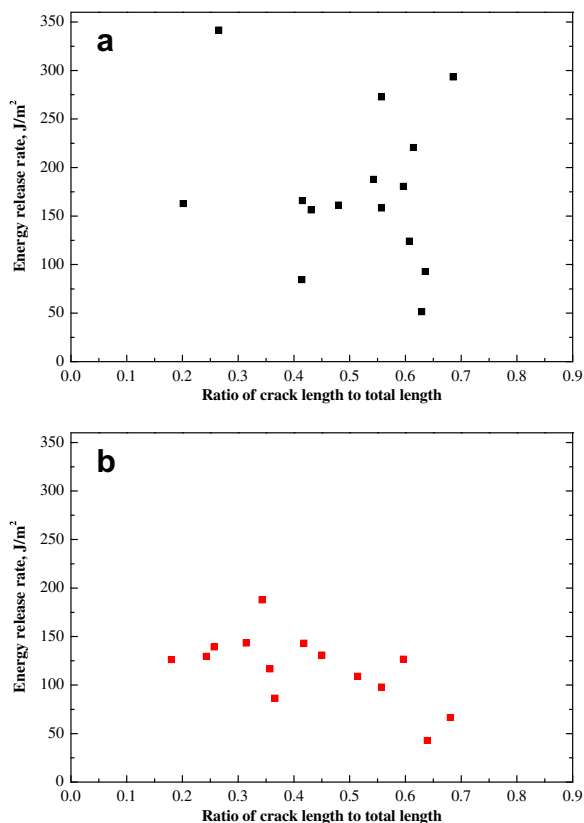


Fig. 10. Adhesive toughness of neat epoxy tested with (a) cracks made by film embedding and (b) cracks by wedge tapping.

using an embedded, non-sticky paper. The adhesive toughness values were plotted in Fig. 10a, corresponding to  $176.9 \pm 79.0 \text{ J/m}^2$ . The large standard deviation may be caused by the varying crack sharpness due to stress concentration and non-uniform paper thickness produced during curing. Fig. 10b contains the adhesive toughness values of neat epoxy-bonded adherends tested with instantly propagated cracks, which led to  $117.6 \pm 34.5 \text{ J/m}^2$ . In comparison with the toughness obtained with embedded cracks, the toughness obtained using instantly propagated cracks is substantially lower, implying that an instantly propagated crack is sufficiently sharp and should be used for adhesive toughness measurement. This is explained in light of the crack tip diameter. An instantly propagated crack tip is around  $1 \mu\text{m}$  in diameter (as demonstrated in Fig. 12 of Ref [13]) compared to the  $40\text{-}\mu\text{m}$ -diameter of an embedded crack using non-sticky paper. The standard deviation of the propagated crack testing is 57.9% less than that of the embedded crack testing, which means that significantly higher reproducibility is produced by the wedge-tapping method.

It is noted that the adhesive toughness  $G_{1c}$  of neat epoxy  $117.6 \text{ J/m}^2$  is lower than the value we reported of  $175.0 \text{ J/m}^2$  using compact tension (CT). This is because CT allows sufficient space to have fracture energy absorbed, while the adhesive toughness testing is conducted within a narrow space of  $0.1 \text{ mm}$  in height where fracture energy cannot be readily consumed.

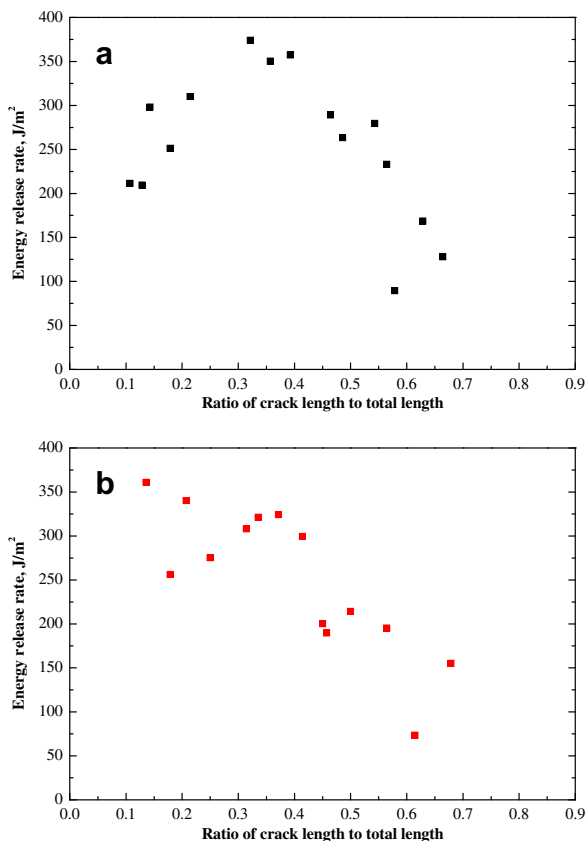


Fig. 11. Adhesive toughness of epoxy/rubber composites tested with (a) cracks made by film-embedding and (b) cracks by wedge-tapping.

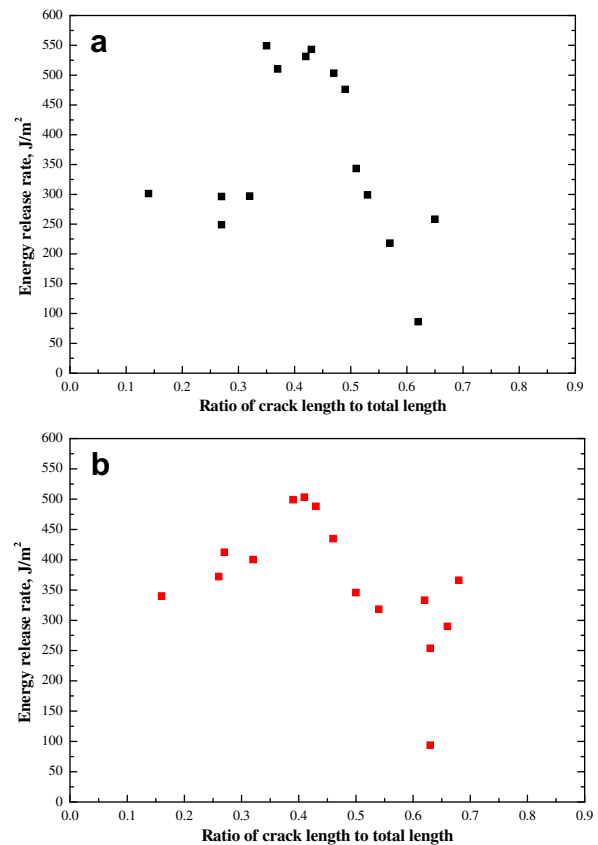


Fig. 12. Adhesive toughness of epoxy/nanosilica composites tested with (a) cracks made by film embedding and (b) cracks by wedge-tapping.

As a conventional toughener for epoxy resins, liquid rubber is dispersed at molecular level when mixed with epoxy and micron-sized particles are formed during curing. 15–20 wt% rubber is sufficient to achieve effective toughening effect, and causes 10–30% loss of the Young's modulus and tensile strength of the matrix. Studies have been conducted on the adhesive toughness of several types of adhesive joints bonded by rubber-modified adhesives [15–18].

Herein, we studied the effect of the crack-producing methods on the adhesive toughness of liquid rubber-toughened epoxy. Fig. 11a contains the toughness values of adherends bonded by 15 wt% liquid rubber-toughened epoxy and tested with embedded cracks, which corresponds to  $254.3 \pm 82.8 \text{ J/m}^2$ . A similar value with similar standard deviation  $250.9 \pm 82.3 \text{ J/m}^2$  was found when these toughened epoxy-bond joints were tested with instantly propagated cracks, as shown in Fig. 11b. This implies that toughened resin is insensitive to the crack sharpness.

Table 1

Comparison of epoxy toughness with different cracks (all samples' crack-total length ratios controlled at  $\sim 0.5$ ).

Methods to create cracks	Embedding razor blade	Embedding Non-stick paper	Tapping
Fracture toughness, $\text{J/m}^2$	$175.5 \pm 90.2$	$185.5 \pm 60.4$	$120.1 \pm 16.8$

**Table 2**

Effect of crack–adherend length ratio on adhesive toughness with instantly propagated cracks.

Materials	Crack–total length ratio	Adhesive toughness, J/m <sup>2</sup>
Neat epoxy	0.1–0.7	117.6 ± 34.5
Neat epoxy	0.2–0.5	134.7 ± 28.5
Epoxy/rubber composite, 15 wt%	0.1–0.7	250.9 ± 82.3
Epoxy/rubber composite, 15 wt%	0.2–0.5	274.7 ± 58.2
Epoxy/silica nanocomposite, 20 wt%	0.1–0.7	363.4 ± 105.7
Epoxy/nanosilica composite, 20 wt%	0.2–0.5	431.9 ± 59.9

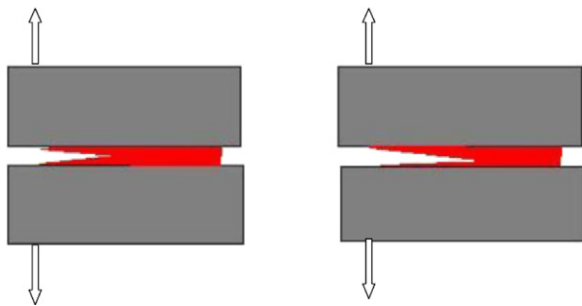
Nanosilica is a well-known stiff toughener for epoxy resins - 20 wt% nanosilica toughened epoxy increases fracture toughness significantly without loss of the Young's modulus and tensile strength of the matrix [13]. Fig. 12a contains the toughness values of adherends bonded by 20 wt% nanosilica-toughened epoxy with embedded cracks, which corresponds to  $364.2 \pm 143.4$  J/m<sup>2</sup>. A similar value but with lower standard deviation  $363.4 \pm 105.7$  was found when these toughened epoxy bonded joints were tested with instantly propagated cracks, as shown in Fig. 12b. The lower standard deviation means that the instantly propagated crack method is more reliable for toughened resins with high stiffness.

Recently, Soon-Ho Yoon proposed a method to create a sharp crack by embedding a razor blade which had been coated with a mold release agent [8]. Thus, we adopted this method to test neat epoxy bonded adherends and compared it with our methods. We prepared a number of adhesive cracks using Yoon's method; two more batches of samples were tested with embedded crack and propagated cracks, respectively. Table 1 shows the adhesive toughness values of neat epoxy tested with these cracks. While the adhesive toughness values collected from the embedded crack methods are similar, both are obviously higher than the value from the propagated crack method.

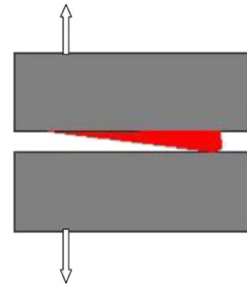
Based on the above analysis, the wedge-tapping method is recommended to create a sufficiently sharp crack for adhesive toughness testing.

### 3.3. Effect of crack length ratio on $G_{Ic}$

In Table 2, the neat epoxy shows 26% reduced standard deviation for the ratios 0.2–0.5 in comparison with the



**Fig. 13.** Cohesive fracture within the adhesive plane (left) or near the interface (right).



**Fig. 14.** Adhesive fracture occurred at the interface.

ratios 0.1–0.7. Smaller standard deviations were also observed for toughened resins. This indicates the importance of setting up a reasonable ratio of crack to adherend length.

### 3.4. Failure of the adhesive joint

Based upon the distribution of adhesive on the fracture surface of adherends, two classes of fracture are well recognized: cohesive fracture and adhesive fracture. As shown in Fig. 13, cohesive fracture is obtained when a crack propagates within the centre plane of the adhesive or near the interface of adhesive and adherend; the fracture surfaces of both adherends are covered with adhesive. In Fig. 14, adhesive (or interfacial) fracture refers to fracture that occurs exactly at the interface between adhesive and adherend; that is, no adhesive is found at the adherend surface when fractured; its appearance implies that the surface treatment of adherend is not good enough. In this study, each adherend fracture surface was carefully examined and no adhesive fracture was found.

## 4. Conclusions

Adhesive testing via loading–unloading cycles is not reliable, regardless of either tapered or non-taper double cantilever beam used. This is due to the plastic deformation caused by the loading–unloading cycles during the propagation of crack (s). Cracks made by a non-sticky insert were not sufficiently sharp and thus produced higher adhesive toughness values with larger standard deviation. By contrast, instantly propagated cracks created by wedge-tapping are sufficiently sharp and yield reproducible, accurate toughness values with lower standard deviation. Toughened adhesive is insensitive to the crack sharpness. The crack length to adherend length ratio of 0.2–0.5 was recommended for adhesive toughness measurement.

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## References

- [1] A.J. Kinloch, *Adhesion and Adhesives: Science and Technology*. Chapman and Hall, London, 1987.
- [2] ISO 25217:2009 Adhesives—Determination of the mode I adhesives fracture energy of structural adhesive joints using double cantilever beam and tapered double cantilever beam specimens.
- [3] ASTM D3433–75 (Reapproved 2005), 'Standard Practice for Fracture Strength in Cleavage of Adhesives in Bonded Joints'.
- [4] R.D. Adams, J.W. Cowap, G. Farquharson, G.M. Margary, D. Vaughn, *Adhesion & Adhesives* 29 (2009) 609.
- [5] T. Yokozeki, T. Ogasawara, T. Aoki, *Composites Science and Technology* 68 (2008) 760.
- [6] A.H. Al-Ghamdi, I.A. Ashcroft, A.D. Corcombe, M.M. Abdel-wahab, *Adhesions* 79 (2003) 1161 (Non).
- [7] M. Imanaka, S. Motohashi, K. Nishi, Y. Nakamura, M. Kimoto, *Adhesion & Adhesives* 29 (2009) 45.
- [8] S.H. Yoon, B.C. Kim, K.H. Lee, D.G. Lee, *Journal of Adhesion Science and Technology* 24 (2010) 429.
- [9] J. Ma, Q. Qi, J. Bayley, X.S. Du, M. Mo, L. Zhang, *Polymer Testing* 26 (2007) 445.
- [10] L. Chen, B.V. Sankar, P.G. Ifju, *Composites Science and Technology* 62 (2002) 1407.
- [11] K. Wang, L. Chen, J. Wu, M.L. Toh, C. He, A.F. Yee, *Macromolecules* 38 (2005) 788.
- [12] Gagliano, J. M.; Frazier, C. E. *Wood Adhesives 2000*, [International Symposium], 7th, United States, 2000; 22–23: 369.
- [13] J. Ma, M. Mo, X.S. Du, P. Rosso, K. Eriedrich, H.C. Kuan, *Polymer* 49 (2008) 3510.
- [14] J. Ma, M. Mo, X.S. Du, S.C. Dai, I. Luck, *Applied Polymer Science* 110 (2008) 304.
- [15] F.J. Guild, A.J. Kinloch, A.C. Taylor, *Material Science* (2010).
- [16] M. Salinas-Ruiz, A. Skordos, I.K. Partridge, *Material Science* 45 (2010) 2633.
- [17] R. Thomas, Y. Ding, Y. He, L. Yang, P. Moldenaers, W. Yang, T. Czigany, S. Thomas, *Polymer* 49 (2008) 278.
- [18] T.H. Hsieh, A.J. Kinloch, K. Masania, J.S. Lee, A.C. Taylor, S. Sprenger, *Material Science* 45 (2010) 1193.