MODE I FRACTURE TOUGHNESS OF ADHESIVELY BONDED JOINTS IN A HIGH TEMPERATURE ENVIRONMENT

M. D. Banea[1], L. F. M. da Silva[2], R. D. S. G. Campilho[3]

[1] Instituto de Engenharia Mecânica (IDMEC),
Rua Dr. Roberto Frias, 4200-465, Porto, Portugal,
E-mail: mbanea@fe.up.pt

[2] Departamento de Engenharia Mecânica,
Faculdade de Engenharia da Universidade do Porto,
Rua Dr. Roberto Frias, 4200-465 Porto, Portugal,
E-mail: lucas@fe.up.pt

[3] Universidade Lusófona do Porto
Rua Augusto Rosa, nº 24
4000-098 Porto, Portugal,
E-mail: raulcampilho@hotmail.com

ABSTRACT

Adhesives used in structural high temperature space and aerospace applications must operate in extreme environments. They need to exhibit high-temperature capabilities in order to maintain their mechanical properties and their structural integrity at the intended service temperature.
As is known, adhesive strength and strain generally show temperature dependence. Similarly, the fracture toughness is expected to show temperature dependence. In order to determine the effect of the temperature on the adhesive fracture toughness of an adhesively bonded joint, pure mode I adhesive fracture toughness tests were performed at high temperatures (100ºC, 150ºC and 200ºC) and at room temperature (22ºC). From these experimental tests, the fracture toughness for the tested temperatures was evaluated for the selected bonded joint system. Experimental results showed a slight increase of the fracture toughness at 100ºC. A drastic decrease in fracture toughness was observed at 200ºC (the T_g of the adhesive was overpassed), but only a slight decrease in fracture toughness at 150ºC was found compared to the room temperature.

KEY WORDS: High temperature adhesives; fracture toughness, temperature tests.

1. INTRODUCTION

There has been a growing requirement in the last years, particularly in the aerospace industry, for adhesives to withstand high temperatures. The adhesives used in structural high temperature space and aerospace applications must operate in extreme environments. Those environments include a wide operating temperature range from cryogenic to 300ºC. These adhesives have to maintain their mechanical properties at the intended service temperature and to maintain their structural integrity (resist thermal breakdown at elevated temperature). Adhesive systems that meet some of these requirements include: epoxies (having high strength and temperature resistance), silicones (excellent sealant for low stress applications, high degree of flexibility and very high temperature resistance), phenolics, polyimides, bismaleimides and ceramic adhesives.

As is known, adhesive strength generally shows temperature dependence. Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [1,2]**.** At high temperatures this is due to the low adhesive strength, while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. Similarly, the fracture toughness is expected to show temperature dependence.

Several investigators addressed the determination of the fracture toughness in tension or shear of thin adhesive layers in adhesively-bonded assemblies, but these studies are often limited to room temperature testing. However, relatively only limited data are available relative to the critical strain energy release rate at low or high temperatures [3,4].

The majority of adhesively-bonded assemblies fracture characterization under pure mode I is performed using the double cantilever beam (DCB) specimen [5,6]. In a fracture mechanics analysis of this specimen, the crack is predicted to propagate when the energy release rate for mode I crack growth (G_I) becomes equal to the toughness of the adhesive or the adhesive's critical energy release rate (G_{Ic}). The main advantages of this test method include its simplicity and the possibility to obtain the fracture toughness mathematically using the beam theory for brittle materials [7].
Several techniques can be used to derive the fracture toughness of structural adhesives from fracture characterization tests. The most common methodologies for analysis are based on Linear-Elastic Fracture Mechanics (LEFM). The Compliance Calibration Method (CCM) is based on the Irwin-Kies equation [8], requiring the calculation of the compliance, C, ($C=\delta/P$, where δ is the displacement and P is the applied load) relative to the crack length during crack growth. The Direct Beam Theory (DBT), based on elementary beam theory [9], and the Corrected Beam Theory (CBT), including the effects of crack tip rotation and deflection [10], are also available within the scope of LEFM. The Compliance-Based Beam Method (CBBM) was recently developed by de Moura *et al*. [11,12] and is based on the crack equivalent concept, depending only on the specimen's compliance during the test.

In this study, the pure mode I fracture toughness of adhesive joints bonded with a high temperature adhesive was measured over a wide range of temperatures. DCB tests were performed at room temperature (RT), 100ºC, 150ºC and 200ºC.

2. EXPERIMENTAL DETAILS

2.1. Adhesive

The adhesive investigated in this study was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase Chemtex (Japan).

A key parameter in the testing of adhesive joints is the glass transition temperature (Tg) of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour. The glass transition temperature (Tg) of the XN1244 adhesive is approximately 160ºC (data provided by supplier).

2.2. Specimen fabrication

Steel substrates were used for the DCB specimens. The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The specimen geometry and the loading are shown in Figure 1.

The bondline thickness was nominally 0.2 mm. Spacers (calibrated steel bars of 0.20 mm) were inserted between the adherends before the application of the adhesive in order to control the bondline thickness. These spacers were removed after the adhesive was cured.

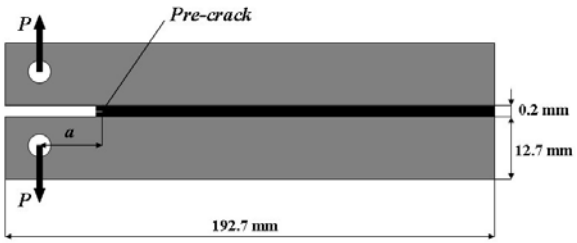


Figure 1. DCB Specimen Geometry.

A sharp pre-crack in the adhesive layer mid-thickness was assured using a razor blade. A mould with spacers for the correct alignment of the adherends was used and is shown in Figure 2. The DCB joints were cured at 140ºC for 1 hour.

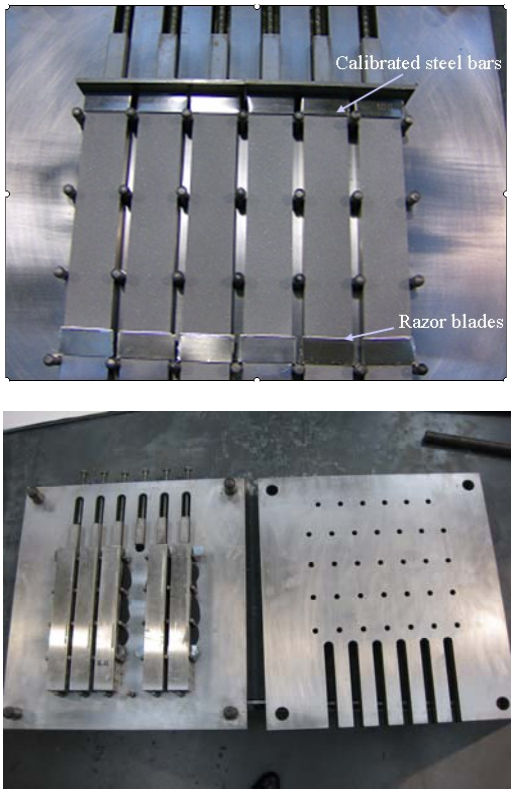


Figure 2. Mould with DCB Specimens.

2.3. Test procedure

The DCB specimens were tested at RT and high temperatures (100ºC, 150ºC and 200ºC) using a universal testing machine Instron® model 8801 (Instron Co., USA), under a constant crosshead rate of 0.5 mm/min. For the high temperature tests, the

environmental chamber of the machine was used to reach the desired test temperatures.

Before the testing was initiated, in order to avoid a blunt crack, all specimens were slightly loaded to ensure 2-3 mm of crack propagation, after which a_0 was measured. The load–displacement (P–δ) curve was registered during the test. Pictures were recorded during the specimens testing with 5 s intervals using a 10 MPixel digital camera.

This procedure allows measuring the crack length during its growth and afterwards collecting the P–δ–a parameters. This was performed correlating the time elapsed since the beginning of each test between the P–δ curve and each picture (the testing time of each P–δ curve point is obtained accurately with the absolute displacement and the established loading rate).

Figure 3 shows a picture of what was recorded during a test that shows the crack tip, allowing the crack length measurement.

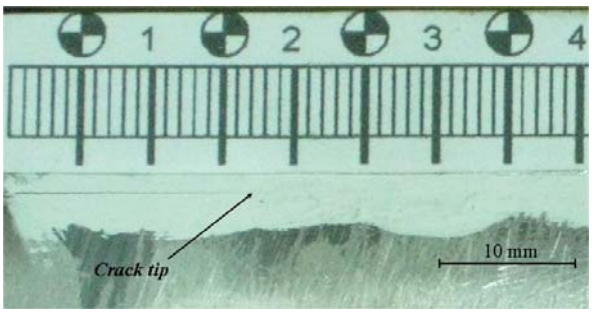


Figure 3. Crack Length Measurement During Propagation.

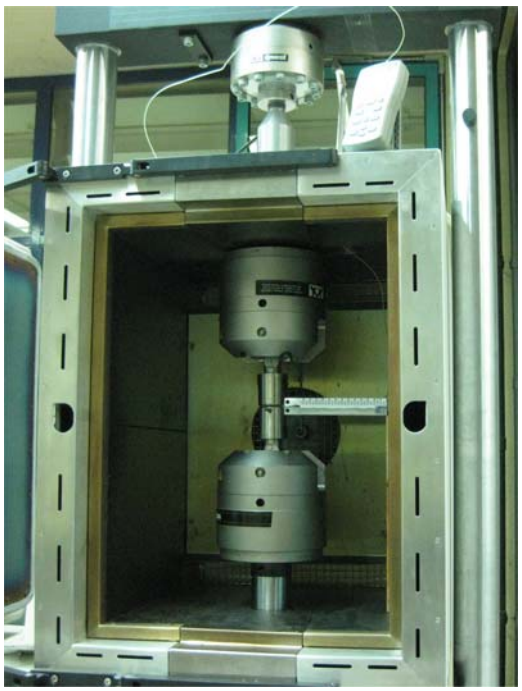


Figure 4. DCB Specimens Set-up.

Four joints were tested to failure at each temperature. The DCB specimens set-up is shown in Figure 4.

A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen's temperature. The tests were always performed after approximately 10 min of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.

2.4. Data analysis

Different methods were employed to evaluate the critical fracture energy in pure mode I, G_{Ic}.

The Compliance Calibration Method (CCM) is based on the Irwin-Kies equation [8]:

$$G_{Ic} = \frac{P^2}{2b}\frac{dC}{da} \quad (1)$$

where P represents the load, b the specimen width and $C=\delta/P$ the compliance.

Cubic polynomials ($C=C_3a_3+C_2a_2+C_1a+C_0$) are used to fit the $C=f(a)$ curves, leading to:

$$G_{Ic} = \frac{P^2}{2b}(3C_3a_3 + 2C_2a_2 + C_1) \quad (2)$$

By the Corrected Beam Theory (CBT), G_{Ic} is obtained using [10]:

$$G_{Ic} = \frac{3P\delta}{2b(a+|\Delta|)} \quad (3)$$

where Δ is a crack length correction for crack tip rotation and deflection.

The Compliance-Based Beam Method (CBBM) was recently developed by de Moura *et al*. [11,12] and is based on the crack equivalent concept, depending only on the specimen's compliance during the test. G_{Ic} can be obtained by the following expression:

$$G_{Ic} = \frac{6P^2}{b^2t_p}\left(\frac{2a_{eq}^2}{t_p^2E_f} + \frac{1}{5G}\right) \quad (4)$$

a_{eq} is an equivalent crack length obtained from the experimental compliance and accounting for the fracture process zone (FPZ) at the crack tip, E_f is a corrected flexural modulus to account for all phenomena affecting the P-δ curve, such as stress concentrations at the crack tip and stiffness variability between specimens, and G is the shear modulus of the adherends.

3. RESULTS AND DISCUSSION

3.1. Determination of G_{Ic} values as a function of temperature

Representative experimental *P–δ* curves of the DCB specimens at each temperature are presented in Figure 5.
The critical fracture energy in mode I was evaluated using the methods presented in Section 2.4.

Table 1 summarizes the test data by presenting G_{Ic} values obtained by each method as well as standard deviation, as a function of temperature. At 100ºC the fracture toughness, G_{Ic}, of the adhesive slightly increased (by approximately 10%). This can be explained by the fact that, as the temperature increases, the strength decreases but the ductility increases giving an additional plastic deformation at the crack tip, hence an increase in toughness. At 150ºC, G_{Ic} slightly decreased, due to the degradation of the mechanical properties of the adhesive induced by the temperature. However, a drastic drop in fracture toughness was observed at 200ºC. This was expected as the testing temperature overpasses the T_g of the adhesive.

Table 1. Fracture toughness G_{Ic} [N/mm] as a function of temperature.

	CBBM	CCM	CBT
RT	0.47 ±0.03	0.43 ± 0.07	0.46 ± 0.04
100ºC	0.52 ± 0.04	0.49 ± 0.06	0.49 ± 0.03
150ºC	0.43 ± 0.06	0.40 ±0.04	0.42 ± 0.03
200ºC	0.08 ± 0.01	0.06 ±0.02	0.07 ± 0.02

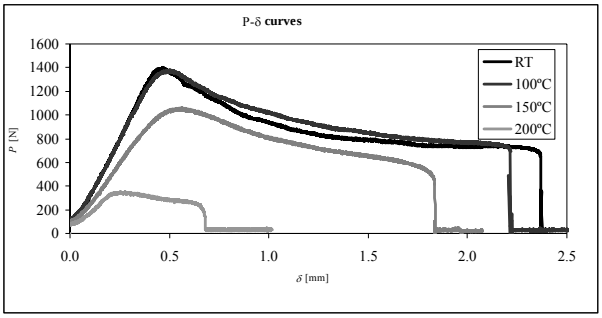


Figure 5. Representative Experimental P–δ Curves of the DCB Specimens as a Function of Temperature.

Experimental *R*-curves obtained by the different methods for one specimen at RT are shown in Figure 6. Similar results were obtained by CBT and CBBM. The CCM presents a slight difference, which is explained by polynomial fitting difficulties. It should be noted that the CBBM *R*-curve is out of phase to the right relatively to the remaining ones, since the equivalent crack used in this method is higher than the real crack length measured during the tests and used in the other two methods (Figure 6).

Experimental *R*-curves obtained by the different methods for one specimen at 100ºC, 150ºC and 200ºC are presented in Figure 7a, b and c.

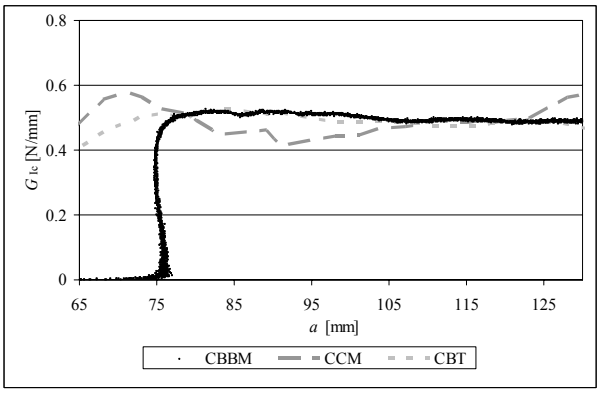
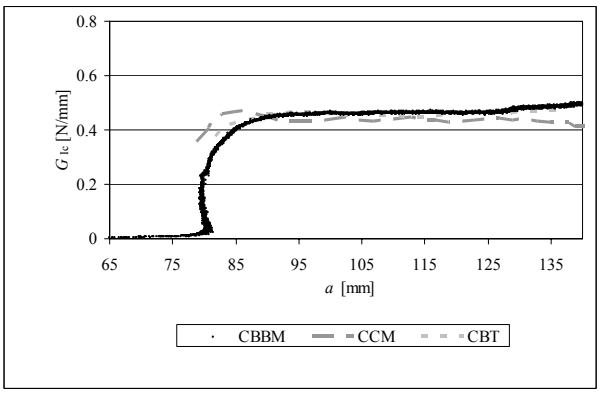
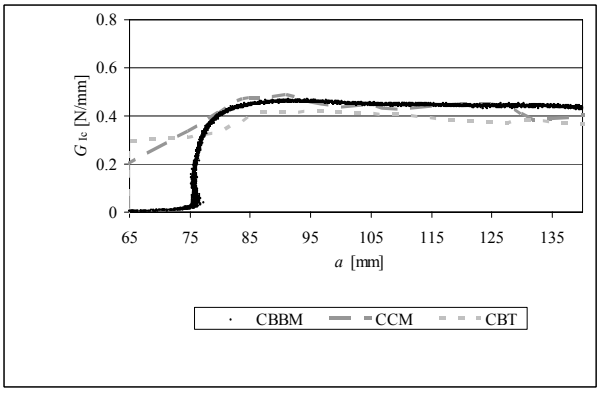


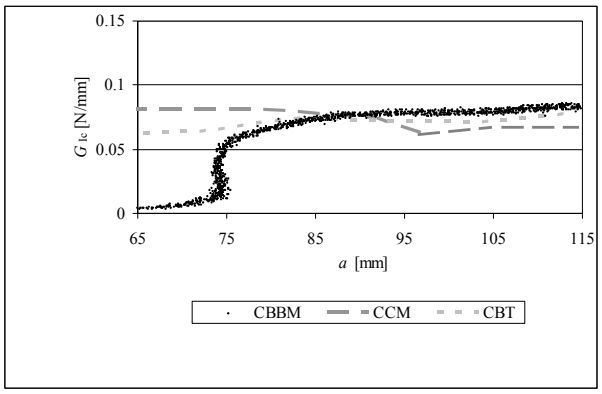
Figure 6. Typical experimental R-curves obtained by the different methods for one specimen at RT.



a)



b)



c)

Figure 7. Experimental R-curves for one specimen at 100ºC (a), 150ºC (b) and 200ºC (c).

3.2. Failure mode

For high strength adhesives there are four typical mechanisms of crack growth, which are represented in Figure 8 [13]. The most representative one is the cohesive failure (a1 and b1), characterized by a proper surface preparation that leads to a stronger interface with the adherends than the cohesive properties of the adhesive. Interfacial failures (c1) are frequent when bonding adherends with low surface energy or as a result of a poor preparation of the bonding surfaces. Alternative crack propagation between interfaces can also appear (d1), due to opening of micro-cracks at opposite interfaces alternatively ahead of the crack tip. The tensile stresses within the bond plane can also play an important role in this behaviour, causing the crack to oscillate within the adhesive layer or alternate from one adherend to the other. This effect is controlled by the T-stress, a non-singular stress that is parallel to the local crack path. If the tensile magnitude of the T-stress is sufficiently large, the crack path is not stable and so will continuously change direction as it propagates [14].

For ductile low strength adhesives, a number of typical failure mechanisms have been reported in the literature: (a2) near-tip void growth and coalescence, (b2) interface debonding near the crack tip, (c2) high triaxiality cavitation ahead of the crack tip and subsequent coalescence, and (d2) interfacial debonding ahead of the crack tip. Schematics of these mechanisms are also shown in Figure 8.
The adhesive studied here belongs to high strength adhesives.

As can be observed in Figure 9 the failure in the DCB specimens was a cohesive failure for all temperatures.

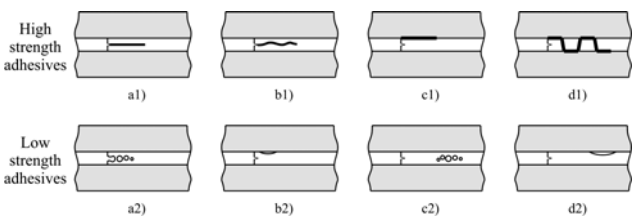
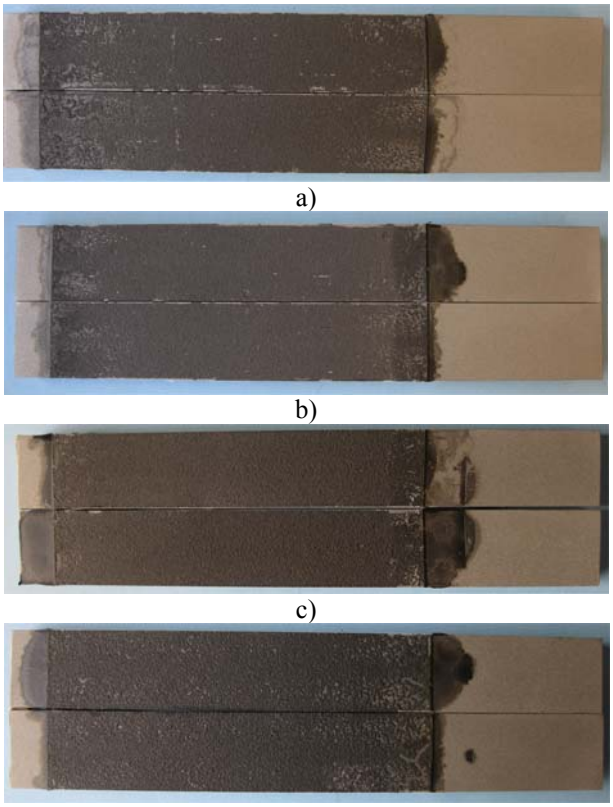


Figure 8. Typical Failure Modes at RT (a), 100ºC (b), 150ºC (c) and 200ºC (d).



a)

b)

c)

d)

Figure 9. Typical Failure Modes at RT (a), 100ºC (b), 150ºC (c) and 200ºC (d).

4. CONCLUSIONS

Mode I adhesive fracture toughness (G_{Ic}) tests were performed at room and high temperatures (100ºC, 150ºC and 200ºC) and the facture toughness G_{Ic} as a function of temperature was obtained for a high temperature epoxy adhesive/steel DCB specimens. At 100ºC the fracture toughness, G_{Ic}, of the adhesive slightly increased (by approximately 10%). This can be explained by the fact that, as the temperature increases, the strength decreases but the ductility increases giving an additional plastic deformation at the crack tip, hence an increase in toughness. At 150ºC, G_{Ic} slightly decreased, probably due to the degradation of the mechanical properties of the adhesive induced by the temperature. However, a drastic drop in fracture toughness was observed at 200ºC, when the testing temperature overpasses the T_g of the adhesive.

ACKNOWLEDGEMENTS

The authors would like to thank the Portuguese Foundation for Science and Technology for supporting the work presented here, through the research project PTDC/EME-PME/67022/2006 and through the individual grant SFRH/BD/61880/2009, and Nagase Chemtex (Japan) for supplying the adhesive.

REFERENCES

[1] Banea D.M., da Silva L.F.M., *Effect of temperature on the Mechanical Properties of the Adhesives for Automotive Industry*, Proc. IMechE, Part L: Journal of Materials: Design and Applications, *in press.*

[2] da Silva, L.F.M. and R.D. Adams, *Measurement of the mechanical properties of structural adhesives in tension and shear over a wide range of temperatures.* Journal of Adhesion Science and Technology, 2005. **19**(2): p. 109-141.

[3] Melcher, R.J. and W.S. Johnson, *Mode I fracture toughness of an adhesively bonded composite-composite joint in a cryogenic environment.* Composites Science and Technology, 2007. **67**(3-4): p. 501-506.

[4] Banea D.M., da Silva L.F.M., Campilho RDSG, *Temperature Dependence of the Fracture Toughness of Adhesively Bonded Joints*, Journal of Adhesion Science and Technology, in press.

[5] Andersson T, Stigh U. *The stress-elongation relation for an adhesive layer loaded in peel using equilibrium of energetic forces.* International Journal of Solids and Structures 2004; 41: p. 413-434.

[6] Ducept F, Davies P, Gamby D. *Mixed mode failure criteria for a glass/epoxy composite and an adhesively bonded composite/composite joint*. International Journal of Adhesion & Adhesives 2000; 20: p. 233-244.

[7] Yoshihara H. *Simple estimation of critical stress intensity factors of wood by tests with double cantilever beam and three-point end-notched flexure*. Holzforschung 2007; 61: p. 182-189.

[8] Kanninen M.F. and Popelar C.H., *Advanced fracture mechanics*, University Press, Oxford (1985).

[9] W. Ding, *Delamination Analysis of Composite Laminates.* Ph.D. Thesis. University of Toronto, Canada (1999).

[10] Robinson, P. and S. Das, *Mode I DCB testing of composite laminates reinforced with z-direction pins: a simple model for the investigation of data reduction strategies.* Engineering Fracture Mechanics, 2004. **71**(3): p. 345-364.

[11] de Moura, M.F.S.F., R.D.S.G. Campilho, and J.P.M. Gonçalves, *Crack equivalent concept applied to the fracture characterization of bonded joints under pure mode I loading.* Composites Science and Technology, 2008. 68(10-11): p. 2224-2230.

[12] de Moura, M.F.S.F., J.P.M. Gonçalves, J.A.G. Chousal, and R.D.S.G. Campilho, *Cohesive and continuum mixed-mode damage models applied to the simulation of the mechanical behaviour of bonded joints.* International Journal of Adhesion and Adhesives, 2008. **28**(8): p. 419-426.

[13] K.M. Liechti, in: *Mechanics of Adhesion,* D. A. Dillard and A. V. Pocius (Eds.), pp. 45-75, Elsevier, Amsterdam (2002).

[14] Chen, B. and D.A. Dillard, *The effect of the T-stress on crack path selection in adhesively bonded joints.* International Journal of Adhesion and Adhesives, 2001. **21**(5): p. 357-368.