Abstract

Adhesively bonded composite joints are exposed to a range of environments in aerospace applications. This paper focuses on the effects of long-term exposure to hot/wet environments on fatigue disbond growth of an adhesively bonded composite joint under Mode I loading. Double cantilever beam (DCB) specimens were manufactured and conditioned to equilibrium at an intermediate and an extreme environment of 70 °C / 95%RH and 80°C / 5% salinity immersion, respectively. The Mode I fatigue tests of these aged specimens were conducted in a humidity chamber at room temperature. Disbond growth of these specimens was monitored using an in-house automated delamination/disbond monitoring system synchronized with the load cycle peak. Quasi-static and fatigue disbond growth rates were characterized for unconditioned and conditioned specimens. The Mode I quasi-static test results showed that the unconditioned specimens maintained virtually the same critical strain energy release rate, \( G_{IC} \), from onset to propagation. However, the conditioned specimens exhibited a large variation in \( G_{IC} \) as the disbond propagated along the specimen, and a much reduced onset \( G_{IC} \) than the propagation values, indicating an important effect of environmental ageing on fracture of adhesively bonded composite specimens. In spite of the large variation in \( G_{IC} \) of the conditioned specimens, a fairly consistent curves were observed for Mode I fatigue disbond growth results where the disbond rates was expressed as a function of fatigue loading defined by the maximum energy release rate, \( G_{\text{max}} \), and a displacement ratio of 0.1. Two test schemes were applied – a continuous one using one set of displacements, and a segmented one with varying displacement sets. It was found that the test schemes had virtually little effects on the results. A Paris relationship was observed for the test region ranging from as low as 0.1 \( G_{IC} \) to close to \( G_{IC} \). No subcritical region or disbond threshold was observed for the test region of this study. Environmental ageing has an apparent effect on disbond growth rates, with a much faster rate being evident at the higher load region. Salt water immersion was shown to cause the most degradation on these adhesively bonded composite joints, resulting in most drastic changes in their fatigue behavior. Subsequent analysis of the failure surfaces revealed a change in failure mode from 100% cohesive to mixed cohesive/adhesive failure that could partially explain the change in fatigue disbond performance of these adhesively bonded composite joints.

1. Introduction

The use of adhesive bonding for part manufacturing and repair of composite structures has become increasingly widespread for aerospace and other industries in recent years [1]. The main issues with the integrity of adhesively bonded composite joints reside in the challenges with materials and process control of surface preparation and bonding, uncertainties in manufacturing and design integration practices, a lack of reliable non-destructive methods for detecting off-nominal strength bonds, commonly called “weak” or “kissing” bonds, and the possibility of undetected in-service damage, etc. Experience has revealed that the presence of process-induced defects and in-service damage has the potential to affect the service life of adhesively bonded joints and in some cases to cause catastrophic failure of bonded sandwich structures in service [2]. Therefore, a better understanding of disbond propagation in an adhesively bonded composite joint is an important
aspect of evaluating the performance of these bonded composite structures.

It is well known that moisture has an effect on the physical, mechanical and chemical properties of polymers. Moisture exists in bulk polymer either in a free or bond states [3]. Free water molecules takes up the free volume among the polymer chains, causing reversible effects of plasticization and reduction in glass transition temperatures of the polymer. Bond water molecule, on the other hand, leads to irreversible damage to the polymer via hydrolysis and chain scission [4]. In addition, the significant difference in moisture absorption between the polymer and the fibres could lead to evolution of localized stress and strain field, leading to early nucleation of micro-cracking [5][6]. For adhesively bonded composite joints, the bondline behavior consistent of substrate/adhesive interface and adhesive becomes further complicated by the surface preparation methods of the substrates, which affects the mechanisms of environmental ageing and degradation [7]. A fundamental understanding of the environmental aging and degradation mechanisms of adhesively bonded composite joints and tools for quantitative prediction of the reduction in performance would enable manufactures to make broader and more efficient use of adhesive bonding. This study focuses on the environmental effects on the Mode I fatigue disbond behavior of the adhesively bonded composite joints.

2. Experimental

2.1 Specimen Design and Fabrication

A modification to the double cantilever beam (DCB) specimen design as per ASTM D 5528-1 [8] was made to conduct fracture toughness tests of an adhesively-bonded joint configuration. The bonded composite specimens were fabricated using two 13-ply of unidirectional continuous carbon fibre-reinforced composite material, Cytec CYCOM 5276-1, bonded with 3M AF163-2K adhesive as shown in Fig. 1. The unidirectional laminates, with an average thickness of 1.8 mm (0.072”), were applied following the cure cycle recommended by the manufacturer. The manufactured CFRP panels were maintained in a vacuum oven at 80 °C until fully dried to ensure there was no pre-bond moisture effect on bond quality. The substrates were abraded with 220 alumina grit and cleaned with nitrogen blast. Two plies of adhesive with a nominal thickness of 0.24 mm (0.0095”) per ply were applied, with a 0.013 mm (0.0005”) thick PTFE film placed between the plies of adhesive at one end of the specimen. Shims and a caul plate were used to achieve a uniform bondline thickness. A two-step cure cycle was applied, with the first at 95 °C and 345 kPa for 20 minutes and the second at 121°C and 345 kPa for 70 minutes. The first step was added to allow excess adhesive to flow out and reduce the overall bondline thickness to values more representative of real applications, where 0.13–0.26 mm thick bondline is often considered optimal [9]. The bonded panels were cut into specimens and hinges were bonded to them with Hysol EA 9360 paste adhesive, as shown in Fig. 1. The final bondline thickness of the test specimens was 0.33 ± 0.07 mm and a loading point is 50.8 mm (2”) away from the centre of the hinge (a0) (see Fig. 1). These DCB specimens were used for both quasi-static and fatigue tests.

![Fig. 1. Schematic of the adhesively-bonded composite specimen](image)

2.2 Environmental Conditioning

In addition to the room temperature ambient (RTA) condition, specimens were conditioned under 70 °C, 95% RH (IVWN), as well as 82°C water immersion of 5% salinity (HISN), as listed in Table 1. These two conditions were selected to simulate an upper limit and an extreme condition that an aircraft structure may be exposed to.

The IVWN specimens were conditioned in an ESPEC PRA-4GP environmental conditioning chamber at the set temperature and humidity level for the entire duration of conditioning. The immersion temperature was maintained in a temperature controlled glass tank heated by heater...
strips bonded to the walls. The water temperature was maintained at ±2 °C of the target temperature. Water salinity was regularly measured and kept at 5% as per ASTM D1141-98 [10]. These DCB specimens were weighed regularly until they reached the equilibrium state as specified in ASTM D5229 [11].

Table 1: Summary of environmental conditions

<table>
<thead>
<tr>
<th>Name</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>RTA</td>
<td>Room Temperature, Ambient humidity</td>
</tr>
<tr>
<td>IVWN</td>
<td>Intermediate temperature (70 °C), Vapour (95% RH.), Fresh Water, No freeze-thaw</td>
</tr>
<tr>
<td>HISN</td>
<td>High temperature (82 °C), Immersion, Salt water, No freeze-thaw cycles</td>
</tr>
</tbody>
</table>

2.3 Fracture Mechanics Approach

The conventional material strength approach is defined by applied stress and material failure strength. In this traditional approach, a design is considered sound when the anticipated applied stress is less than the material failure strength. This approach may be adequate for brittle fracture by imposing a safety factor on stress, combined with minimal tensile elongation requirements on the material, but is not adequate for cases where flaws or damage propagate in a progressive manner, leading to failure of the structure [12].

The energy-based fracture mechanics approach defines that failure occurs when the energy available for crack growth exceeds the resistance of the material. Strain energy release rate, one form of fracture toughness is defined to be the loss of energy, $dU$, in the test specimen per unit of specimen width for an infinitesimal increase in disbond length, $da$, for a disbond growing under a constant displacement (see Eqn. 1).

$$ G = -\frac{1}{b} \frac{dU}{da} \tag{1} $$

For the Mode I double cantilever beam (DCB) case, $G_I$ can be expressed in Eqn. [2] based on modified beam theory (MBT) method [8]. Failure occurs when $G_I \geq G_{IC}$, the critical strain energy release rate.

$$ G_I = \frac{3P\delta}{2ba} \tag{2} $$

where, $P$ is the load, $\delta$ is the displacement of the loading point.

The Mode I DCB test was carried out in the displacement control mode, based on the test procedure for Mode I fracture toughness outlined in the ASTM D5528-01 and ASTM D6115-97 [13] for static and fatigue tests, respectively. Load $P$, and displacement $\delta$ were measured experimentally using a calibrated load frame, while disbond length $a$ was determined by an in-house automated disbond measurement system (ADMS) which allowed for disbond tip image capturing from optical camera system at the peak load or peak displacement. It was assumed that no further disbond growth would take place beyond the point of peak displacement in that cycle, thus enabling the $G_{imax}$ value of each peak to be calculated.

To determine the maximum cyclic displacement $\delta_{imax}$ for any given max strain energy release rate $G_{imax}$ in a fatigue test, the values of critical strain release rate, $G_{ic}$, and critical displacement, $\delta_c$, for the same disbond length were first determined. These critical values were obtained either from the static DCB test results of specimens from the same batch. For each desired $G_{imax}$ value, maximum cyclic displacement $\delta_{max}$ for testing was estimated from the relationship:

$$ \frac{\delta_{max}^2}{\delta_c^2} = \frac{G_{imax}}{G_{ic}} \tag{3} $$

where both $\delta_c$ and $G_{ic}$ were determined from static tests.

2.4 Mode I Quasi-static Fracture Toughness Testing

The Mode I DCB quasi-static tests were conducted according to ASTM D5528 in laboratory conditions, at a temperature and humidity level of 22±5 °C and 50±20% RH, respectively. The test utilized a MTS 858 Tabletop hydraulic test frame, with a calibrated 250N or a 500N load cell with 1% load accuracy of measurements. The data acquisition station used TestStar II Station Manager v3.4B to record the load and crosshead displacement during testing.
The edge of the specimens was painted with a thin layer of white coating, with a scale of 0.05 mm graduation bonded to the lower half of the specimen. The prepared specimen was placed in between the grips of the load frame as seen in Fig. 2. The disbond lengths were recorded when disbond length reached the pre-determined values based on visual observation using travelling microscope. Each of these quasi-static fracture tests typically took a few minutes to perform. Therefore, moisture loss during the test was assumed to be negligible.

Fig. 2. Gripped specimen in the load frame

2.5 Mode I Fatigue Disbond Growth Testing

Mode I fatigue fracture toughness tests at the lab environment for the RTA specimens were conducted in a similar manner to the quasi-static testing. Once the displacement reached the target maximum value, fatigue loading was applied to the specimen at a set frequency. Time, cycle count, load and displacement data were collected by the load frame control software. It is not an easy task to measure disbond length during a fatigue test. Direct and indirect disbond length measurement methods have been developed, such as visual observation, crack gauge, dynamic and quasi-static compliance calibration [14][15]. In this study, an automatic delamination/disbond monitoring system (ADMS) was developed to capture the images of the disbond tip at cycle peak value.

All specimens were conditioned in the environmental chambers at their respective environment until the equilibrium state was reached where no further weight change was observed. The fatigue tests were conducted at the room temperature. As shown in Fig. 3, an environmental chamber made from transparent Plexiglas was used to maintain consistent humidity level in the conditioned specimens throughout the entire duration. Humid air generated from a cold-dispensing atomizer humidifier passed through the chamber during the test. The level of humid air intake was adjusted to ensure minimal weight change in the specimens. A camera for disbond tip image capturing was mounted in front of the environmental chamber. To enhance image quality, heater strips set at 40°C were placed on the side wall of the heater to remove water condensation in the viewing window area during disbond monitoring.

The calculation of Mode I strain energy release rate, $G_{IC}$, was based on the measurement of specimen compliance and disbond length, while specimen compliance can be directly obtained from load frame measurements.

Fig. 3. Close-up view of humidity chamber with humidity intake attached and coupon mounted

2.6 Disbond Monitoring

To overcome the issues posed by the aforementioned methods such as high cost, limited data points, or data reliability, an automatic delamination/disbond monitoring system (ADMS) was developed in this study to measure disbond growth directly in a continuous basis. This was conducted by acquiring images of disbond tip positions synchronized with peak loads during a fatigue test, or acquiring images along with load or displacement measurements from the frame during a static test. The frequency of image capturing varied from 1 frame per 100 cycles up to 20 frames per second, subject to the rate of
disbond growth, and the image acquisition frequency can be tailored to match the rate of disbond growth.

The ADMS consists of a digital camera, a 3-axis travelling base, and a Labview based-control system to analyse frame signals and to send a trigger for the camera operation. These images (see Fig. 4 for an example) were manually examined in post-test analysis using UTHSCA ImageTool version 3.0 [1]. This non-contact system allows for frequent disbond length measurement as often as desired in an uninterrupted manner, ensuring data quality and accuracy. The flexibility that the system offers in terms of data density and potential for real-time determination of disbond length for test control, make it an attractive technique for crack/disbond monitoring for both metallic and composite fatigue testing.

![Image](image_url)

Fig. 4. A sample image acquired from the automatic delamination/disbond monitoring system (ADMS)

2.7 Fatigue Test Matrix

In order to evaluate hygrothermal ageing effects on fatigue disbond growth of adhesive bonded composite joints under Mode I loading, a total of six DCB specimens were tested for each group specimen subjected to either the RTA, IVWN or the HISN condition, as shown in Table 2. The displacement control mode with R-ratio of 0.1 was employed in this study. For the displacement-controlled fatigue testing, the maximum strain energy release rate decreased as disbond propagation. Each group of six specimens were tested using two different test schemes – three following a continuous scheme and the other three a segmented test scheme. Under the continuous test scheme, the same maximum and minimum displacements were set to achieve an initial target \( G_{\text{imax}}/G_{\text{ic}} \) value of 1.5 and maintained throughout the test. Due to the large displacement required to reach the targeted \( G_{\text{imax}}/G_{\text{ic}} \), the test frequency range was limited to maintain the sinusoidal loads of 14.7 kN (3.3 kips) hydraulic frame. Under the segmented scheme, the tests started with an initial target \( G_{\text{imax}}/G_{\text{ic}} \) of 0.3 followed by higher displacement values to achieve the targeted \( G_{\text{imax}}/G_{\text{ic}} \) value of 1.5. The smaller crosshead displacement for the slow growth region made it possible to apply a higher frequency such as 2.5 Hz, leading to a much reduced test time.

Those tests using a continuous scheme typically terminated when \( G_{\text{imax}}/G_{\text{ic}} \) dropped to 0.3, and the segment tests resulted in a runout \( G_{\text{imax}}/G_{\text{ic}} \) as low as 0.15 for the RTA specimens. For conditioned specimens, the \( G_{\text{imax}}/G_{\text{ic}} \) lower limit was slightly higher due to the observed slower crack growth rates at the low load region.

Table 2: Test matrix for fatigue propagation test

<table>
<thead>
<tr>
<th>Test Scheme</th>
<th>Specimen type</th>
<th>Displacement</th>
<th>( G_{\text{imax}}/G_{\text{ic}} )</th>
<th>No. of repeats</th>
<th>R-ratio</th>
<th>Frequency (Hz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Continuous</td>
<td>RTA, IVWN</td>
<td>Constant</td>
<td>1.5-1.0</td>
<td>3</td>
<td>0.1</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>HISN</td>
<td>1.0-0.3</td>
<td>3</td>
<td>0.1</td>
<td>1.5</td>
<td></td>
</tr>
<tr>
<td>Segmented</td>
<td>RTA, IVWN</td>
<td>Low</td>
<td>0.3-runout</td>
<td>3</td>
<td>0.1</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>HISN</td>
<td>High</td>
<td>1.5-1.0</td>
<td>3</td>
<td>0.1</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.0-0.3</td>
<td>3</td>
<td>0.1</td>
<td>1.5</td>
<td></td>
</tr>
</tbody>
</table>

3. Experimental Results and Discussion

3.1 Critical Strain Energy Release Rates

The critical energy release rate, \( G_{\text{ic}} \), represents the threshold of resistance to disbond growth under Mode I quasi-static loading. The values of onset and propagation strain energy release rates were generated in a static test, where pre-cracking was performed following ASTM D5528 to remove the effect of the Teflon insert.
The onset and propagation $G_{IC}$ values were obtained for the specimens that were subjected to three different environmental conditions – RTA, IVWN, and HISN. Five specimens were tested for each condition. All the $G_{IC}$ values were calculated using the MMB method according to ASTM D5528, and 5% offset or max load was used for onset $G_{IC}$ calculation, where the 5% offset was defined to be the intersection of the curve with 5% increase in compliance from the linear portion of the $P$–$\delta$ curve with this curve.

Experimental results of the Mode I fracture toughness in Fig. 5 showed that environments played an important effect on onset and propagation $G_{IC}$ of the adhesively bonded composite joints. It can be seen that $G_{IC}$ of the RTA specimens were largely consistent throughout the whole test region, with little difference between the onset and propagation values. However, large variations were observed for the fracture toughness of both groups of conditioned specimens. It was found that for the studied adhesively bonded joints, the onset $G_{IC}$ of the conditioned specimens was significantly lower than the propagation values, indicating a reduced resistance to disbond onset due to environmental degradation. The average of the onset $G_{IC}$ values were found to be 2795, 2356, and 1917 J/m$^2$ for specimens conditioned at RTA, IVWN and HISN, respectively.

As per ASTM D5528, the load and displacement values were recorded at 1 mm disbond length increments for the first 5 mm, followed by load and displacement data at every 5 mm, until the disbond propagated at least for 45 mm from the tip, and again at every 1 mm increment of crack growth for the last 5 mm of disbond. This varying data frequency allowed for an improved data capture of change in $G_{IC}$ in the region where $G_{IC}$ typically varied the most during propagation. A weighted average $G_{IC}$ during propagation was obtained taking into account the different increments of disbond growth, as shown in:

$$G_{IC} = \frac{1}{(a_{n+1} - a_1)} \sum_{i=1}^{n} G_{IC,i} \times (a_{i+1} - a_i) \quad (4)$$

Where $a_i$ is the $i$th measured disbond length and $n$ is the total number of disbond length measurements during propagation. $G_{IC,i}$ is the calculated strain energy release rate at the $i$th measurement.

Using this weighted approach, an average value of propagation $G_{IC}$ for RTA specimen was calculated to be 2847 J/m$^2$, while the values for specimens conditioned at IVWN and HISN were found to be 2854 and 2498 J/m$^2$, respectively. This indicates that extended exposure to infusion at 70 °C, 95% RH significantly affected the ability of the joints to resist disbond onset; however, this resistance to further disbond propagation was found to be similar to that of an unconditioned specimens. In comparison, it was shown that the bonded joints subjected to 82°C salt water immersion had the greatest reduction in fracture toughness for both onset and propagation. Due to the large scatter of the data, a larger number of tests for the conditioned specimens are recommended to generate statistically important values of $G_{IC}$.

### 3.2 Fatigue Disbond Growth

As shown in Fig. 6, the two test protocols gave the identical fatigue disbond growth curves for the adhesively bonded composite joints under Mode I, with excellent repeatability. The test results also showed that, the disbond growth rates followed a power law relationship to the maximum strain energy release rate in the test range of 0.1 to 1.0 $G_{IC}$. The disbond growth beyond the critical strain energy release rate was characterized by an unstable growth trend. No disbond threshold was observed for the test range.
3.3 Effects of Environmental Ageing

Fig. 7 and Fig. 8 show the Mode I fatigue disbond growth of the adhesively bonded composite DCB joints that had been conditioned at 70 °C, 95% RH and 82 °C salt water, respectively, until they reached the equilibrium state. Similar trends were observed for the conditioned specimen, with a power relationship between the growth rate and the maximum strain energy release rate until approached or exceeded the $G_{I_{\text{max}}}$ values obtained from the quasi-static fracture toughness tests. Similarly, no subcritical region was observed within the test region.

As compared to the unconditioned specimens, there was a slightly larger variation in the disbond growth curve from specimen to specimen. Nevertheless, the data were considered to be fairly consistent given the extraordinarily high variations observed in the $G_{I_{\text{c}}}$ onset and propagation curves of the conditioned specimens under quasi-static loading, as shown in Fig. 5.

A comparison of the disbond growth rates are plotted in Fig. 9. On the log-log scale, the RTA specimen has the lowest slope, indicating the least dependency of the growth rate on $G_{I_{\text{max}}}$. In comparison, disbond growth rate of the HISN specimens are the most sensitive to the change in Mode I loading. It is also interesting to observe that while environmental ageing led to a faster disbond growth rate in the higher load or $G_{I_{\text{max}}}$ region, and a slower rate in the lower load region. Generally, immersion at a higher temperature led to a higher level of ageing and degradation of bondline fracture toughness of these composite joints.

Fig. 6. Mode I fatigue disbond growth of unconditioned (RTA) adhesively bonded composite joints.

Fig. 7. Mode I fatigue disbond growth of adhesively bonded composite joints subjected to 70°C, 95% RH (IVWN).

Fig. 8. Mode I fatigue disbond growth of adhesively bonded composite joints subjected to 82 °C, immersion of 5% salinity (HISN).
Fig. 9. Comparison of the steady fatigue disbond growth region of adhesively bonded composite Joints after long-term exposure to RTA, IVWH and HISN conditions.

A subsequent failure mode study of the specimens revealed that while unconditioned specimens failed 100% cohesively, the environmentally degraded specimens had a mixed cohesive/adhesive failure, as shown in Fig. 10. Such change illustrates the complex failure mechanisms at the bondline, where environmental ageing affect adhesive properties, but it also changes the failure mechanisms. While the hygrothermal effects on material behaviors such as strength and viscoelasticity can be obtained using established testing and modelling methods, there is lack of fundamental understanding of such change in failure mechanisms and predictive tools for conducting a quantitative assessments.

It is conceivable that there were several competing factors associated with environmental ageing in play – viscoelasticity of polymers, change in material properties and strengths, as well as a change in failure mode. While enhanced plasticity as a result from high temperature and humidity may lead to stress relaxation, the reduction in strength of the materials as well as possibly early nucleation at the adhesive/substrate interface result in weakening of bonded joints.

Fig. 10 Failure surfaces of Mode I fatigue specimens (a) RTA with 100% cohesive, (b) IVWN with mixed and (c) HISN specimens with mixed cohesive/adhesive failure mode.

4. Conclusions

This study focuses on the environmental ageing and degradation on fatigue disbond growth of adhesively bonded composite joints under Mode I loading. It was found that disbond growth rates for all specimens followed a Paris behaviour to $G_{i,max}$ in the region of $G_{i,max} < G_{ic}$. No sub-critical region, where disbond rate would drop to zero, was observed for these tests. The test results showed that environmental degradation affected the fatigue life of the bonded joints in the highly loaded cases in a different manner from that in the lower loading cases. As compared to the unconditioned specimens, faster Mode I fatigue disbond growth rates were observed when subjected to higher loads, and slower rates at the low load region for those environmentally aged specimens. The environmental condition of 82 °C, immersion in 5% salinity led to a more severe degradation than that of
70 °C, 95% RH. The subsequent failure analysis showed that unconditioned specimens failed cohesively, whereas a mixed cohesive/adhesive failure mode was observed for conditioned specimens. It is conceivable that there were several competing factors associated with environmental ageing in play – viscoelasticity of polymers, change in material properties and strengths, as well as a change in failure mode. The Mode I quasi-static fracture tests also showed that the critical strain energy release rates for unconditioned specimens were fairly constant. Onset $G_{IC}$ of these unconditioned specimens was found to be the same as the propagation value. In comparison, a large scatter in $G_{IC}$ for onset and propagation was observed for the conditioned specimens, and $G_{IC}$ at onset was found to be significantly lower than the values at propagation. Such large variations in $G_{IC}$ in conditioned specimens however, did not seem to lead to high scatter in fatigue disbond curves.

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References