THE EFFECT OF PREPREG STORAGE HUMIDITY ON CO-CURED COMPOSITE JOINTS

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Introduction

The increasing use of composite materials in the aerospace industry has driven a need for a greater understanding of bonded composite joints.

There are generally two types of composite joint used in the aerospace industry; secondary bonded joints and co-cured joints. Secondary bonded joints are produced by bonding two cured composite laminates together with an adhesive. However, when composites and adhesives are used to manufacture large parts in the aerospace industry, it is often convenient to co-cure the two materials at the same time. This helps to reduce the high costs associated with autoclave curing and also to reduce processing time.

However, despite the apparent advantages, co-curing is not without its drawbacks. Any moisture stored in the composite material prior to co-curing is released during the cure cycle and has a negative effect on the joint. This can also result in interfacial failure. A way around this problem is to either dry the composite material prior to curing or to engineer the composite surface using a variety of surface treatments to promote adhesion, such as an atmospheric pressure plasma treatment [1]. The former option will be investigated in this work.

The effects of moisture on the fracture performance of secondary bonded composite joints is well publicised. Moisture can be introduced into the composite laminate prior to [2] or after [3] secondary bonding. The moisture can plasticize the adhesive and reduce the glass transition temperature of the adhesive [4]. However, compared to secondary bonded joints, relatively little work has been carried out on co-cured joints.

In the present work, the effect of the level of moisture in the composite prepreg prior to co-curing will be examined.

Experimental

Materials & Manufacture of Specimens

The materials used in this study include an aerospace grade thermoset composite prepreg and an aerospace grade film adhesive manufactured and supplied by Cytec Engineered Materials.

The co-cured joints were manufactured in-house at University College Dublin using a press-clave and vacuum bagging procedure. The joints consisted of 20 ply of unidirectional prepreg with a layer of film adhesive between the 10th and 11th ply. A 12 micron thick Teflon sheet was used as a crack starter during the layup. The press-clave was heated up to 180 ºC over 2 hrs and then held at 180 ºC for 2 hrs under a constant pressure of 80 psi as per the manufacturers guidelines.

Once cured, specimens were cut to a size of 25mm x 170mm using a diamond grinding disc. The specimens were approximately 5.5mm thick with an average bondline thickness of 0.2mm.

LEFM Tests

Three linear elastic fracture mechanics (LEFM) based tests were conducted to determine the fracture toughness of the co-cured joints. The tests employed were a mode I double cantilever beam (DCB) test [5], a mixed mode I+II asymmetric double cantilever beam (ADCB) test [6] and a mode II end loaded split (ELS) test [7]. All tests were conducted at a constant crosshead displacement rate of 1 mm/min at room temperature on a screw-driven Hounsfield 50K tensile test machine. The propagation values for G_C were calculated using corrected beam theory (CBT) from the following equations.

DCB: $$G_{IC} = \frac{3P\delta}{2B\left(a + |\Delta_I|\right)} \cdot \frac{F}{N}, \qquad (1)$$

ADCB: $$G_{I/IIC} = G_{IC}^{M} + G_{IIC}^{M}, \qquad (2)$$

where: $$G_{IC}^{M} = \frac{3P^2\left(a + |\Delta_I|\right)^2}{B^2 E h^3} . F, \qquad (3a)$$

and: $$G_{IIC}^{M} = \frac{9P^2\left(a + |\Delta_{II}|\right)^2}{4B^2 E h^3} . F, \qquad (3b)$$

ELS: $$G_{IIC} = \frac{9P^2\left(a + |\Delta_{II}|\right)^2}{4B^2 E h^3} . F, \qquad (4)$$

where P is the applied load, δ the crosshead displacement, B the width of the specimen, h the half-thickness of the specimen, $\Delta_{I/II}$ the crack length correction term, F the large

displacement correction factor, N the load block correction factor and E the flexural modulus of the substrate.

Humidity Control

Prepreg sheets were stored in a humidity controlled environment for 1 week prior to co-curing. The humidity level was controlled using saturated salt solutions. Four humidity levels were examined; 11, 43, 75 & 98% RH using lithium chloride, potassium chloride, sodium chloride & potassium sulphate saturated respectively.

DTA & TGA

A Scientific Rheometric STA1500 was used to investigate the thermal and thermogravimetric properties of the adhesive and prepreg respectively.

TGA was used to determine the moisture weight loss of the prepreg material over the course of a cure cycle.

DTA was used to determine the glass transition temperature, T_G, of adhesive scrapings taken from the fracture surface of the LEFM test specimens. The adhesive was heated from 25 ºC to 250 ºC at a rate of 10 ºC/min. T_G was defined as the mid-point of the transition.

Results and Discussion

TGA Results of Prepreg

Moisture is stored in the composite prepreg in one of two states; free water that can be removed by drying or heating the prepreg to 100 ºC and bound water that forms multiple hydrogen bonds with the thermoset polymer network and can not be removed by drying but can be removed by heating to 180 ºC [8]. Figure 1 shows a typical TGA trace for a prepreg material. Note the two distinct regions where weight loss occurs which correspond to the two states of stored moisture.

Figure 2 compares TGA traces for the as-received prepreg with ones that have been stored at various humidity levels. It can be seen that as humidity level increases, the weight loss associated with free water also increases.

Figure 3 shows the quantities of water lost due to free and bound water. There is a strong correlation between the humidity level and percentage weight of free water. However, the level of bound water does not appear to change with humidity level.

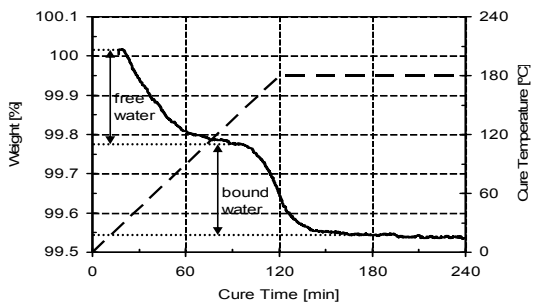


Figure 1: Typical TGA trace of prepreg over the course of a cure cycle.

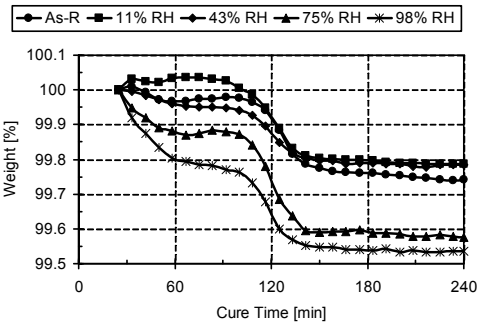


Figure 2: TGA traces of as-received (As-R) prepreg and after storing at various humidity levels for 1 week.

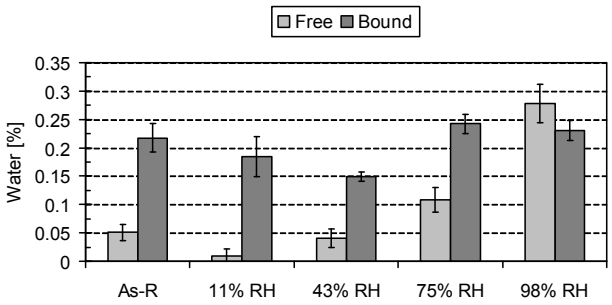


Figure 3: Free and bound water in prepreg after 1 week at various humidity levels. Results are the mean of 3 samples and error bars represent the standard deviation.

LEFM Test Results

Mean propagation values of G_C obtained from DCB, ADCB and ELS tests are shown in Figure 4a, 4b & 4c respectively for co-cured joints prepared using the as-received (As-R) and humidity conditioned prepreg.

Comparing the joint behaviour with that of the as-received material, it can be seen that the fracture toughness increases slightly for prepreg samples stored at 11% RH as free water content is reduced (see Figure 3). However, failure remained interfacial for all tests.

As the storage humidity level increased above 11%, there was a reduction in the fracture toughness for all three test types. Higher humidity levels seemed to have a more pronounced effect on mode II fracture toughness.

Co-cured joints prepared using the as-received materials resulted in interfacial failure for all tests. It was observed that, as humidity level increased, the locus of failure changed from interfacial to cohesive. This was due to the moisture plasticizing the adhesive and reducing its cohesive strength. While the ADCB test showed a decreasing trend in fracture toughness above 11% RH, $G_{I/IIC}$ never fell below that of the as-received material (Fig 4b). This is likely the result of a change in the locus of failure.

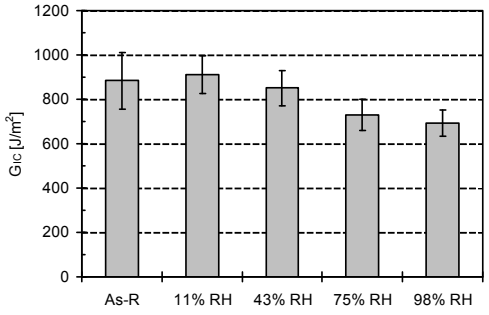
Samples of adhesive were scraped from the resulting fracture surfaces and analyzed using DTA to determine the glass transition temperature. Figure 5 shows the results of the analysis. As humidity level rises, T_G of the adhesive is reduced indicating that it has been plasticized.

Conclusions and Future Work

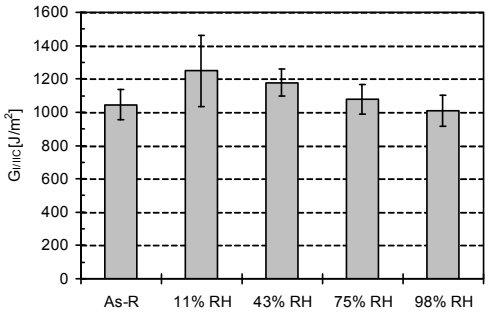
The main conclusions from the present work are:

- Co-cured composite joints resulted in interfacial failure caused by moisture stored in prepreg.
- Moisture was stored in the prepreg as free and bound water.
- As storage humidity level was increased, the level of free water increased. The level of bound water did not change with humidity level.
- There was a reduction in the mode I and mode II fracture toughness as humidity level increased.
- A corresponding decrease in the glass transition temperature of the adhesive was also observed.
- The locus of failure changed from interfacial to cohesive at high humidity levels for all tests.
- There was a strong correlation between free water content, fracture toughness and T_G.

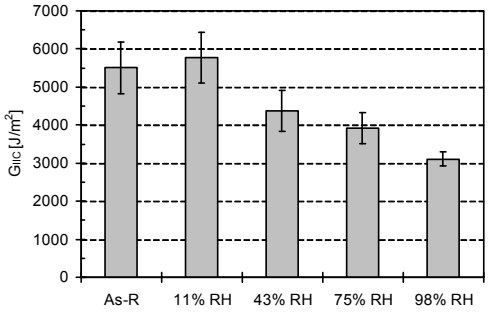
Future work is concerned with investigating the morphology of the fracture surface using scanning electron microscopy in an attempt to relate damage mechanism to fracture toughness. Current preliminary investigations on the effect of atmospheric plasma coatings on the adhesion characteristics of the co-cure composite joint are also being conducted.



(a) Mode I DCB



(b) Mixed-Mode ADCB



(c) Mode II ELS

Figure 4: G_C results of co-cured joints produced using as-received material and humidity conditioned prepreg.

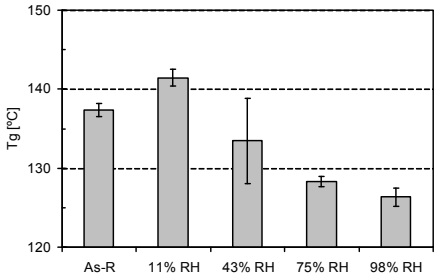


Figure 5: T_G of adhesive taken from fracture surface of LEFM test specimens.

Acknowledgements

The authors would like to gratefully acknowledge the financial support of the Irish Research Council for Science, Engineering & Technology (IRCSET) and Cytec Engineered Materials (CEM). The materials supplied by CEM are also gratefully acknowledged.

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