EFFECT OF HYGROTHERMAL CYCLES ON MECHANICAL PERFORMANCE OF COMPOSITE ADHESIVELY BONDED JOINTS

Sonia Rincon Urbina, David Ayre, Hamed Yazdani Nezhad*

Enhanced Composites & Structures Centre, School of Aerospace, Transport & Manufacturing, Cranfield University, United Kingdom
*Corresponding author: h.yazdani-nezhad@cranfield.ac.uk, Web Page: http://www.cranfield.ac.uk

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Abstract

This paper numerically and experimentally studied mechanical performance of composite adhesively bonded single-lap joints in the presence of hygrothermal cycles, under static tensile loading. Joint performance was predicted by the development of a coupled experimental-numerical approach based on cohesive zone modelling.

Composite adherends of aerospace grade carbon fibre-reinforced Hexply® M21/T800 pre-impregnated plies, bonded using a 25mm × 25mm bond overlap. Bond interface was exposed to cyclic moisture and temperature loads by introduction of 2mm sharp cracks at joint runouts. Pre-cracked joint specimens were subjected to hygrothermal cycles in environmental chamber under conditions representative of aircraft operational cycles.

Testing proved that joint degradation occurred with increased cycle numbers. Strength reduced by 42% under static load after 714 cycles compared to unaged joints. Degradation accelerated in the initial 84 cycles, but was reduced for higher cycles attributed to adhesive bulk moisture saturation. Moisture diffusion parameters were characterised for both adhesive and composite subjected to hygrothermal cycles. Adhesive reached moisture saturation level of 1.54%wt, while composite laminate was 0.68%wt. In both cases, moisture diffusion followed Fick's second law. Displacement-diffusion analysis determined effect of moisture on elasticity of adhesive. This analysis plus the single-lap test data were coupled to develop degradation parameters required for CZM, demonstrating an 87% accuracy at 714 hygrothermal cycles.

1. Introduction

The Aircraft Industry continuously faces repair issues of composite structures; engineers have to decide what type of joint is able to maintain the integrity for a given time and environment, encouraging the necessity to know about bonded joint characteristics over a long time. Adhesive joints have shown significant advantages compared to traditional mechanical fastenin [1-4], because of their lower weight, reduced stress concentration, and high specific stiffness and strength that improve the structural performance [5].

However, the use of bonded joints in primary structures and repairs have been restricted due to Airworthiness Authority's concerns about aviation-safety issues related to a lack of knowledge about long-term durability, difficulty with quality assurance (e.g. via non-destructive inspection), and limited standardisation of the manufacturing process and repair techniques [6]. Therefore, a study for joint strength prediction in terms of life and strength will provide confidence when using adhesively bonded-joints, instead of the mechanically fastened joints which reduce mechanical properties in tension by 40% to 60%, and in compression by 15% [7].

The main goal of this research is to evaluate the structural strength of an adhesive joint in a composite laminate system made of carbon fibre-reinforced polymer (CFRP) as it is exposed to a harsh environmental condition, and to develop an efficient finite element model to predict the joint behaviour.

The degradation environment in these laboratory examinations has involved mechanical and hygrothermal effects including temperature, moisture and mechanical stress.

2 Materials and experiments

Composite single lap joints bonded with FM94 film adhesive and bulk adhesive specimens were used to perform the experiments.

2.1 Materials, geometry and manufacture

A composite panel (2 mm thickness) was manufactured in an autoclave, with a quasi-isotropic stacking sequence of $[0°/45°/90°/-45°]_S$, from aerospace grade unidirectional prepreg Hexcel T800/M21. Prior to bonding, the cured panel was cut into smaller panels having dimensions of 300mm × 100mm . The panels were then dried out in an oven to remove moisture trapped on the panels' surfaces. A bonding fixture was used to ensure bonding overlap dimensions (25mm bond length) and accurate alignment of the joints. The panels were then mutually bonded by secondary bonding procedure and peel ply surface treatment. A debonding strip of polytetrafluoroethylene (PTFE) with 0.1 mm thickness and 2 mm width was laid in the free ends of bonded joints in order to induce a pre-existing nearly zero-thickness bond defect (Figure 1). These were used to avoid a fibre Tear Failure (FT) [8], and thus ensure consistent failure results. Also balancing end tabs were bonded on the opposite faces of the panels to minimise the eccentricity of the applied tensile load [9]. The bonded panels were then cured in the autoclave at 120°C and 0.28 MPa pressure. The bonded plates were cut to produce single lap joints (SLJ) of 25mm width, 175 lengths and 25 mm overlap (25mm×25mm bond area), according to ASTM D5868-01 (reapproved 2014).

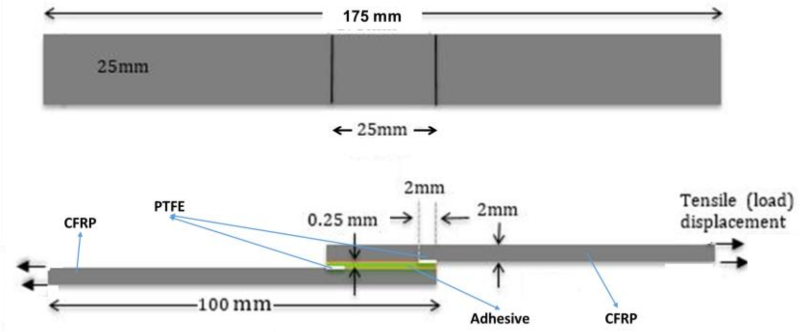

Figure 1. Geometry of the single lap joint specimen

To characterise the moisture diffusion and mechanical properties of the materials (composite and adhesive), specimens of the composite adherend and bulk adhesive were prepared. To obtain the bulk adhesive specimens of 1 mm thickness, a lay-up of four layers of FM94 was used, each having 0.25 mm thickness, and cured. The bulk adhesive specimens were cut with dimensions of 150mm×10mm for mechanical testing. The CFRP T800/M21 specimens with dimensions of 50mm×50mm were manufactured for moisture diffusion characterisation.

2.2 Experimental method

In order to investigate the effect of hygrothermal ageing cycles on the materials' static response, the bulk adhesive, CFRP T800/M21, and the SLJ specimes were exposed to the cycles using a controlled environmental chamber. To simulate the fligth operation hygrothermal conditions, multiple cycles of heating up to 70 °C, with 85 % relative humidity holding for four hours, then cooling down to -20 °C holding for two hours were carried out continually with each cycle taking eight hours. the specimens were conditioned up to a maximum of 714 cycles, then they were removed to be tested under static load.

2.2.1 Gravimetric test

The moisture uptake by the composite and bulk adhesive during the cycles was determined to calculate the diffusion coefficient, D, and the moisture concentration, M, at saturation as recommended by ASTMD5229/D5229M-14. To measure the moisture gained, all pre-dried specimens were conditioned in an environmental chamber under the hygrothermal cycle. The samples were removed periodically from the environmental chamber and kept in a sealed bag as they were being moved to the balance area. Samples were wiped with cloth to remove the moisture from the surface, and then they were weighed immediately on a balance with an accuracy of 0.01g. After weighing, the samples were enclosed within the bag again and moved to the enviromental chamber, these measurements were repeated over a period of time until the moisture percentage achieved the equilibrium. The equilibrium or saturation is reached when the average moisture content changes by less than 0.02 % over at three consecutive time intervals [10].

2.2.2 Mechanical test

The bulk adhesives tensile testing followed the standard EN ISO 527-1:2012 [11], three tensile samples of each set were tested. Unaged specimens (0 cycle conditioned) were tested to identify the initial condition as reference specimens. Aged specimens were conditioned at different periods in cycles (weeks): 84 (four weeks) and 168 (eight weeks).
The SLJ tests were carried out according to standard test method ASTM D5868-01, 2001. The universal machine, Instron 5500R with 30 kN load cell, was used for testing. A laser extensometer was employed to measure the deformation of the joint. The test ran until failure with a displacement rate of 1mm/min. The specimens were removed and tested at the following number of cycles (weeks): noungh (0), 42 (two),84 (four),168 (eight), 252 (12 weeks), and 714 (34 weeks).

3 Results and discussion

3.1 Moisture absorption

The results obtained in this study showed that the moisture absorption in both FM94 adhesive and T800/M21 laminate, exposed to hygrothermal cycles (simulating flight operations), follows the Fick´s second law [12]. It showed a dynamic behaviour with the highest absorption rate during hot-wet stages (non-flight). FM94 reached the moisture saturation of 1.54%wt. at 120 hours (168 hygrothermal cycles) of exposure, while T800/M21 achieved moisture saturation at 0.68%wt. at 672 hours. This is due to lower diffusion in different fiber direction of the laminate, while the diffusion of the adhesive behaved as isotropic material.
Moisture diffusion in both FM94 bulk and composite T800/M21 exposed to the hygrothermal cycles have been fitted to a Fick's second law expression as $F = -D\frac{\partial c}{\partial x}$ [13]. Changes in the diffusion rate can be observed in Figure 2 between hot-wet and cold-dry conditions; these changes have evidenced diffusion dependence on temperature. Temperature reductions results in a drop in diffusivity and the maximum moisture content. However, the moisture diffusion profile is driven at the hot-wet conditions. According to this approach, commercial aircraft industries have affirmed that the moisture content depends mainly on the ground relative humidity during non-flight operations [14].

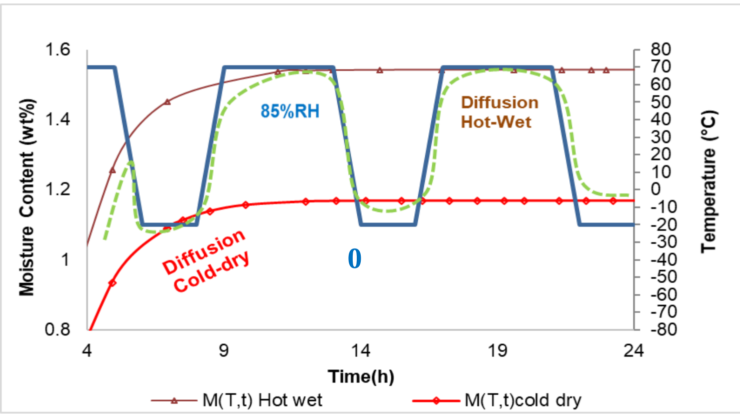

Figure 2. Schematic cyclic diffusion FM94

3.2 Bulk tensile test results

The Young's modulus of the bulk adhesive has not shown a significant change after taking the maximum moisture content of 1.54 %. The elastic modulus is noticeably smaller than 3000 MPa reported by Zavatta [15], but it agrees with 1750 MPa reported by Roh, H.S [16].

3.3 Joint strength

The joint strength decreased gradually with increasing number of hygrothermal cycles. The main reduction occurred at 42 cycles, the strength reduced by 23% compared to that of the unaged specimens. The decreasing trend continued until 84 cycles, and after that the strength reduction was levelled off. After 714 cyles, the loss in the joint strength was 42 %.
The maximum failure load was 6.58 kN (Figure 3) for the unaged samples. It was 52 % lower than the expected maximum load. The lower ultimate load is attributed to the influence of multiple factors such as moisture entrapment on the adhesive bond, surface preparation and the pre-crack tip in the joint.

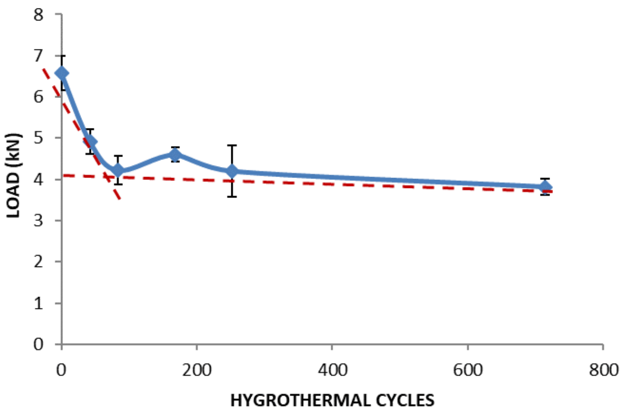

Figure 3. Average joint failure load under hygrothemal cycles

3.3.1 Numerical modeling and discussion

A methodology that couples experimental data with a numerical modelling approach has been proposed. A diffusion moisture model was established to determine the effect of the moisture concentration on the elastic modulus of the bulk adhesive through a thermal-displacement analysis in Abaqus. The moisture characterisation results shown in section 3.1 were used as input for performing the bulk finite element (FE) analysis. An adhesively bonded single lap joint (SLJ) degradation was modelled based on cohesive law. . The degraded parameters of SLJ were determined based on the joint´s experiments and analysis of the bulk adhesive.

3.4 Moisture analysis

The numerical model of the bulk adhesive was carried out to predict the elastic modulus degraded as a function of the moisture cycles. For the diffusion analysis of the bulk adhesive the concentrations of humidity found in the experiments were employed, section 3.1, those evaluating the three different number of hygrothermal cycles**:** 42 (1.49 %wt), 84 (1.51 %wt), 168 (1.54 %wt).
An analogy between heat transfer law (Fourier's Law) and moisture diffusion law (Fick's Law) [17] was used to perform a 3D moisture displacement analysis; density and specific heat can be taken as a unit as proposed by [18]. The coefficient of moisture expansion (CME) used was 0.0016/ $wt(\%)$ [19] [20], and the diffusion coefficient taken from the experimental results.
The results of the bulk adhesive moisture-diffusion displacement analysis showed a reduction on the E modulus by 20 % after 42 hygrothermal cycles (1.49 % moisture content), and with the increment of moisture the elasticity remained nearly constant until saturation (1.54 % moisture content). Later, the reduction trend levelled off exhibiting slight changes reaching a value of 2 414 MPa at the maximum moisture concentration.

These findings, contrary to the FM94 tensile experimental results, are consistent with other researchers [18, 21] and with the plasticity variation observed in the joints post failure evaluation. For that reason, the E degraded values were used to calculate the joint stiffness degraded as a parameter in the CZM.

3.5 SLJ Cohesive zone model

The SLJ degradation was predicted using a bi-linear CZM to simulate the composite-adhesive interface mechanical response. SLJ geometry configuration and boundary conditions are shown in Figure 1. Adherends were modelled using 3D continuum elements with reduced integration (C3D8R).
The adhesive was modelled using 3D cohesive elements layer (COH3D) to represent cohesive crack propagation path at the bond [22]. Two millimetres length were reduced at each end of bondline elements to simulate the crack origin. A quadratic nominal stress criterion (QUADS-in Abaqus) was used [23] [24]
Fracture energy was chosen as damage evolutions criterion with a linear power behaviour [25, 26]. To calibrate the parameters, the fracture energy was taken from experimental studies and the cohesive traction was calibrated via comparison of simulations and the experiment results[25].
The stiffness parameters (Kn and $Ks = Kt$) were obtained by dividing the Young´s modulus (E) and Shear modulus (G) by the adhesive thickness [24, 27], e.g. $Kn = \frac{E}{t}$; and $= Kt = \frac{G}{t}$. Values of G_{I_C}=1.7 and G_{II_C} =2.5 N/mm have been reported by Zavatta [15].
The adhesion failure mode showed that the adhesive/adherend interface strength was less than that estimated. A approach suggested by Belnoue *et al. (adhesion failure approach)* [28], to model adhesion failure with CZM was implemented in this study to calibrate the unaged CZM parameters. Calibration of CZM parameters based on unaged bulk experiments were conducted for modelling adhesive bonded joints [27, 29].
The model predicted a failure load of 6.8kN for unconditioned joints, in full agreement with the actual data i.e. 3.8 % greater than the average experimental peak load. Later, a second crack initiated at the opposite end and both cracks propagated in direction X. The failure occurred when both cracks met in the middle of the overlap.

3.6 Degradation parameters

For the aged joints, the properties of the adhesive/adherend interface, the cohesive parameters (traction and fracture energy) were reduced as the degradation rate showed in the joints after the hygrothermal cycles, represented in the Figure 4. The degraded stiffness, K_{deg} for each number of cycles was obtained by dividing the E numerical modulus of adhesive bulk results by the adhesive thicknesses.
To determine the degraded traction as a function of the hygrothermal cycles, the gradient of reduction (from the lap shear strength results) was used as a factor of degradation. Hence, the empirical equation (3-1) was defined to predict the traction in tension (${T_n}^{Deg}$) as a function of number of cycles (h).

$${T_n}^{Deg} = {T_n}^0 \ (2e^{-6} h^2 - 0.0018h + 0.8626) \qquad \textbf{(3-1)}$$

As fracture energies were reduced proportionally to the cohesive strength reduction, then the fracture energy mode I was obtained using the equation (3-2), where the δ^f calculated for unconditioned state remained constant for all ageing conditions.

$${G_{I_C}}^{deg} = \frac{{T_n}^{deg}}{\delta^f} \qquad \textbf{(3-2)}$$

The outcomes showed good correlation with the experiments predicting the unaged joint strength with an error of 4%.However, the predicted unaged joint stiffness had an error of 20% compared with the experiments, this may occur due to the fact that the E modulus employed was reported in the literature and was found to be higher than that viewed in the experiments.

The prediction of the degraded strength was accurate and kept within the margin of standard deviation up to 252 cycles. Nonetheless, at 714 hygrothermal cycles the model over predicted the strength by an error of 13 %. This unexpected increment in the predicted failure could be attributed to a deviation offered by the empirical equation used to calibrate the parameters.

Table 1. Properties of aged adhesive and cohesive used in the CZM of SLJ

ADHESIVE				COHESIVE			
Hygrothermal Cycles	E (MPa)	Kn (N/mm³)	Ks=Kt	Tn (MPa)	$Ts = Tt$	G_{I_c} (N/mm)	G_{II}
0	3000	12000	4444	36	15	1.7	1.75
42	2432	9728	3602	28	12	1.32	1.36
84	2422	9688	3588	26	11	1.23	1.26
168	2414	9656	3576	22	9	1.04	1.07
252	2414	9656	3576	19	8	0.9	0.92
714	2414	9656	3576	21	8.8	0.99	1.02

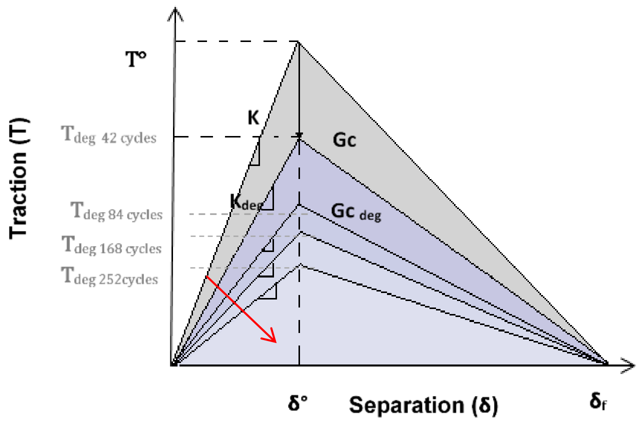

Figure 4. Schematic degradation process for bilinear traction-separation curve

Table 2. Load-displacement results validation SLJ

CONDITION	STIFFNESS		FAILURE LOAD		DISPLACEMENT	
Hygrothermal Cycles	FEM (N/mm³)	Exp	FEM (kN)	Exp Avg.	FEM (mm)	Exp Avg
0	68.71	54.40	6.84	6.58	0.24	0.16
42	67.05	54.20	5.59	4.92	0.19	0.15
84	66.46	51.23	5.27	4.22	0.18	0.16
168	65.47	57.22	4.56	4.60	0.16	1.14
252	64.44	52.29	4.02	4.20	0.15	0.15
714	64.5	52.16	4.34	3.82	0.15	0.13

4 Conclusions

The results of this investigation showed the extent of damage and strength reduction of the composite SLJ, providing valuable data to validate the predicted cohesive zone modelling. A combined experimental-numerical approach was employed to predict the strength and crack propagation in aged joints. The model included harsh and realistic conditions, such as mechanical damage (induced crack initiator) and exposition to humidity and temperature cycles. The key findings were:

- Moisture absorption of both FM94 adhesive and T800/M21 laminated exposed to hygrothermal cycles (flight operations) in line with Fick´s Second Law, with a dynamic behaviour that showed the highest absorption rate during hot-wet stages (on the ground or non-flight).

- The E modulus and the tensile strength of the adhesive FM94 did not evidence changes after the exposure to hygrothermal cycles. Whereas the shear strength of the SLJ showed reductions of about 42 % after 714 hygrothermal cycles compared with the control specimen.
- The hygrothermal environment is most likely to affect the adhesive/adherend interface of the SLJ specimen and 'saturation' was achieved after 84 hygrothermal cycles. Under static load the reduction reached a stabilized condition after 84 hygrothermal cycles
- The numerical diffusion moisture-displacement analysis results are consistent with experimental observations reported by others [19, 20, 29]), but do not agree with the experimental findings of this work - the elasticity dropped by 20 % after 1.4 % moisture uptake (42 hygrothermal cycles).
- This conservative CZM model allows a prediction of the behaviour of SLJ to be made with an accuracy of 4 % for zero hygrothermal cycles, 1 % for 252 hygrothermal cycles, and 13 % for 714 hygrothermal cycles.

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