The strength prediction of adhesive single lap joints exposed to long term loading in a hostile environment

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Abstract. This work is concerned with investigating the residual static strength of adhesively bonded joints after long-term exposure to a combined mechanical-hygro-thermal environment. Associated experimental data are also reported. The degradation process of the joints was modelled using a fully-coupled approach, with the moisture concentration affecting the stress distribution and the stress state affecting the moisture diffusion analyses simultaneously. A bilinear cohesive zone model was then used to implement the progressive damage FE analysis of the quasi-statically loaded joints following the ageing phase. This model is degraded using the damage factors (creep strain and moisture uptake) accumulated over the ageing process and calibrated against the experimental results from static tests on the bulk adhesive. Predicted and experimentally-measured quasi-static responses for the aged adhesive joints were found to be in good agreement.

1. Introduction

Adhesives have been widely used in the automotive, aerospace and construction industries to replace the conventional joining techniques. [1]. The lack of accurate strength prediction of adhesively bonded joints exposed to a long-term hot-humid environment inhibits a more wide-spread application of adhesive bonding. A reliable strength prediction model is essential to reduce the amount of expensive and time-consuming durability testing at the design stage [2-6].

The diffusion of water ingress in adhesives has been modelled extensively by Fick's second law [7-9]. The deleterious effect of water on the integrity of adhesive joints has been investigated by Gledhill and Kinloch. [10]. The measured strength of their joints saturated in distilled water decreased considerably and higher immersion temperatures led to more rapid degradation. It is worth noting that no further degradation was observed after saturation. Brewis et al. [11] found that plasticisation occurred in the adhesive they studied which, in the form of the single lap joint, was exposed in a hot-humid environment for up to 2500 hours.

The residual stress in adhesive joints caused by adhesive swelling and the mismatched

thermal expansion coefficients may relax due to creep. Thus it is important to determine the viscoelastic properties of the adhesive to provide reliable prediction of the adhesive behaviour during long-term degradation. Peretz and Weitsman [12] presented creep tests on FM73 bulk adhesive to investigate the viscoelastic characteristics at elevated temperatures. A temperature range of 30 °C to 60 °C was considered and the results showed that it is essential to include thermal and viscous effects in the characterization scheme. Jurf and Vinson [13] studied the effect of moisture on the viscoelastic shear properties of FM73M and FM300M and found that moisture can enhance the creep rate significantly.

Crocombe [14] studied the environmental degradation of the static strength of adhesively bonded structures assuming both interfacial and cohesive failures based on experiments and FEM simulations of joints bonded by FM1000 adhesive and immersed in water. Sugiman et al. [15-16] modelled the moisture diffusion process in the adhesive layer of adhesively bonded joints under both static and fatigue loading and immersed in deionised water for up to 2 years based on experimental bulk adhesive data. The joint strength and fatigue resistance were found to be degraded by moisture ingress after long term immersion. A similar trend was found when investigating the effects of cyclic thermal loading on adhesive single lap joints by Hu et al. [17]. Results revealed that a cyclic temperature environment decreased joint strength significantly initially and the degradation rate decreased as exposure time increased.

Progressive environmental damage has been used to predict the residual strength of degraded adhesive joints using a continuum damage model [5, 18] or a cohesive zone model (CZM) [4, 19-21]. However, fully-coupled situations where stress and moisture uptake occur simultaneously have not been modelled. In this work, prior to numerical simulation, experimental work was carried out to measure: a) the stress dependency of moisture uptake in the bulk adhesive; b) the moisture dependent creep compliance of the bulk adhesive; c) the stress-strain curves of the bulk adhesive degraded by creep and by moisture and d) the residual strength of degraded adhesively bonded single lap joint (SLJ) specimens. Numerical modelling was then performed in two steps: 1) to characterise the long-term ageing process in adhesively bonded joints under combined thermal-hygro-mechanical loading conditions using a fully-coupled methodology and 2) to simulate the quasi-static tensile residual strength of the aged adhesive joints employing a CZM model which used a bilinear traction-separation law.

2. Experimental methods

2.1 Specimen manufacturing

Experimental studies have been carried out on both bulk adhesive specimens and aluminium single lap joints bonded with FM73 (Cytec®, New Jersey, USA) film adhesive with a nominal thickness of 0.18 mm. A plate of bulk adhesive has been made by stacking and curing nine layers of FM73 film and then machining into dogbone specimens with overall length, gauge length and gauge width of 65mm, 30mm and 5mm respectively. The thickness of the bulk specimens was maintained at 1 mm using steel spacers. The film was cured at 120 °C for 1 hour as recommended by the manufacturer [22]. Further details related to the manufacturing process can be found elsewhere [15, 23-24].

Two 2024-T3 aluminium adherends (45 mm in length, 4.7 mm in thickness and 3 mm in width) bonded with FM73 adhesive constitute the single lap joints (10 mm overlap length and 0.2 mm adhesive layer thickness) investigated in this work. Surface preparation was applied to the aluminium substrate by Airbus before being bonded, first using chromic acid etching (CAE) followed by phosphoric acid anodising (PAA) and then by applying the corrosion inhibiting primer BR127. The detailed specification for the actual treatments are not available but typical specifications are available in the literature [25, 26] for CAE and PAA respectively. The joint manufacturing was carried out in an environmentally controlled room to avoid excessive dust particles. The two aluminium substrates were bonded together in a spring-loaded jig and a pressure of 0.3 MPa was applied to the overlap area. To maintain an adhesive thickness of 0.2 mm, steel spacers with thickness of 4.9 mm were used. The curing procedure was the same as for the bulk adhesive described above.

2.2 Bulk adhesive ageing

To investigate the moisture diffusion, creep and thermal and swelling expansion behaviour in the bulk adhesive, the bulk specimens were immersed (some loaded and some unloaded) in deionised water at 50°C for 6 months. A spring-loaded rig, shown schematically in Fig. 1, was designed to provide the required loading levels (both unloaded and at 25% of the static failure load) for the bulk specimens. The length of the spring was periodically re-adjusted during the loading period to provide essentially a constant load on the specimen.

The weight of the specimens was measured periodically over a period of time until saturation was achieved. This method is known as the gravimetric method which was used to obtain the coefficients of moisture diffusion and equilibrium moisture uptake. Further details of this method can be found elsewhere [27]. At the same time as the weighing procedure, the swelling of the bulk adhesive (assumed to be isotropic) was determined by measuring the specimen thickness using a micrometer [26, 28].

The CTEs of FM73 adhesive and aluminium alloy 2024-T3 were required to determine the thermal stresses induced in cooling from the curing temperature of 120 °C. The CTE for aluminium alloy (Al) 2024-T3 can be found elsewhere to be $2.36 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$ [29]. This material was used as a reference material when determining the CTE of the adhesive. Strain gauges were bonded on both the aluminium and the adhesive and were placed in an oven to measure the strain variation with increased temperature. The relationship between the CTEs for the adhesive and the reference material (Al 2024-T3) can be deduced from Eq. 1 [30].

$$\alpha_A - \alpha_R = \frac{(\varepsilon_{T/O/(G/S)} - \varepsilon_{T/O/(G/R)})}{\Delta T} \quad (1)$$

here, α_A and α_R are CTEs for the adhesive and reference materials, $\varepsilon_{T/O/(G/S)}$ and $\varepsilon_{T/O/(G/R)}$ are the strain outputs for adhesive and reference materials and ΔT is the temperature change from the initial reference temperature to the current value.

Measurement of the extension of the bulk adhesive, strained in the spring loaded frame

(Fig. 1) provided the creep behaviour of the bulk adhesive at 25% of the unaged static failure load tested at room temperature (RT). These specimens were initially dry when immersed and loaded and absorbed increasing amounts of water as the test progressed. Thus they change from a dry to a saturated specimen. The creep data obtained from these tests were supplemented with previous creep data on specimens that were dry and specimens that were pre-saturated before loading. These data were available for a range of load levels. A power-law creep model was used in the subsequent simulation procedure as expressed in Eq. 2 [31].

$$\dot{\epsilon} = Aq^n t^m \quad (2)$$

here, $\dot{\epsilon}$ represents the creep strain rate, q is the von Mises equivalent stress, t is the time and A , n and m are coefficients based on a fitting procedure to the experimental data.

Fig. 1. Loading jigs for the bulk adhesive degradation test.

2.3 Single lap joint ageing

The SLJs were constrained in spring-loaded jigs and immersed in deionised water at 50 °C under different loading levels (12.0% and 17.5% of the dry joint strength tested at RT) to investigate the effect of stress on the joint ageing response. The joint extension caused by creep was determined by periodic measurement of the displacement of the pre-compressed spring.

2.4 Quasi-static tensile testing

Quasi-static tensile testing has also been carried out at RT on both unaged and aged bulk adhesive and SLJ specimens. The testing was conducted using an Instron 6025 universal testing machine and the elongation of the bulk specimen was measured with an extensometer. Moisture dependent stress-strain (load-displacement for the SLJs) curves were obtained from the resulting data.

3. Experimental results

Based on the experimental methods described above, the following material properties of FM73 adhesive were obtained. The CTE and CHE for the adhesive were found to be respectively $8.00 \times 10^{-5} \text{ }^{\circ}\text{C}^{-1}$ and $0.00463 (\%m_w)^{-1}$ (m_w being the mass of water in the adhesive). The equilibrium moisture uptake and moisture dependent Young's modulus of the adhesive are given in Table 1. It can be seen that the Young's modulus dropped by about 15% after

saturation. The Young's modulus and CTE used for the 2024-T3 aluminium alloy are 70 GPa and $2.36 \times 10^{-5} \text{ }^{\circ}\text{C}^{-1}$ respectively and the plastic response in the aluminium alloy was also included as presented in Table 2.

Fickian diffusion was used to characterise the moisture ingress in the adhesive and the dependency of the moisture diffusion on the stress level is shown in Table 3 [19]. It can be seen that the rate of moisture diffusion is significantly accelerated by the applied stress. Further, the stress is seen to increase the saturated mass uptake of water by over 25%. The increase in both parameters might be explained by the increase in the free volume of water in the bulk adhesive under loading [26].

The creep parameters, based on the power-law model discussed in Section 2.2, are shown in Table 4. A curve fitting procedure was carried out to determine these parameters based on the measured extensions from dry, pre-saturated and gradually saturated bulk specimens [31]. Good agreement was achieved comparing the experimental and numerical creep strain-time curves for the gradually saturated bulk adhesive as presented in Fig. 2. It is worth noting that a two-phase (primary and secondary) creep model was used to simulate both a parabolic curve in the first phase (with time-hardening effect) and a linear curve in the second phase (without time-hardening effect), which jointly provide a better fit to the experimental data. The secondary creep rate data was obtained from the specimens illustrated in Fig. 1. These were initially dry but became progressively saturated. By the time the secondary creep rate data was measured these specimens would have been effectively saturated. In the Step 1 FE modelling, discussed later, creep parameters at a point in the material are interpolated between the dry and saturated values depending on the moisture level at that point in the adhesive. The same secondary rate has been used for both conditions. This is reasonable because, as with the experimental data, by the time the transition creep strain has been reached the adhesive will generally be in a saturated condition. Good correlation was found between the predicted and experimental creep behaviours in the SLJs as presented elsewhere [32].

Based on the quasi-static testing at RT of the aged bulk adhesive specimens, the moisture and creep dependent adhesive Young's modulus and failure strength were obtained as presented in Table 5. It can be seen that moisture had a significant effect on the degradation of adhesive properties leading to the decrease of 13.9% and 16.9% in Young's modulus and failure strength respectively in unloaded condition, while applying sustained load in hot-wet environment led to a further reduction of 9.5% and 5.3% in Young's modulus and failure strength respectively. However, applying sustained load on its own only resulted in a degradation of 7.0% and 1.1% in Young's modulus and failure strength respectively.

Table 1 Young's modulus for FM73 at various moisture concentrations.

Environment	Moisture content / %	Young's modulus / MPa
Dry / 50°C	0	1650
Saturation / 50°C	3.75	1400

Table 2 Plastic properties of the aluminium alloy 2024-T3 [23].

Stress / MPa	Plastic strain / %
300	0.000
330	0.003
370	0.015
420	0.043
440	0.100

Table 3 Fickian diffusion parameters for FM73 adhesive immersed in deionised water.

Stress / MPa	Temperature / °C	Saturation content / % m_w	Diffusion coefficient / m^2/s
0	50	2.95	5.21×10^{-13}
11.75	50	3.75	7.18×10^{-13}

Table 4 Creep parameters for FM73 at 50 °C (force / N, length / mm, time / s, stress / MPa).

Phase	Condition	A	n	m
Primary	Dry	5.774E-010	4.75	-0.4764
Primary	Saturated	1.398E-009	4.75	-0.397
Secondary	Both	2.956E-013	4.75	0

Fig. 2. The numerical and experimental creep strain-time curves at 50°C and 25% of the static failure load (tested at RT) for FM73 bulk adhesive.

Table 5 Material parameters for the aged FM73 adhesive tested at RT.

Degradation environment	Moisture content / %	Creep strain / %	Young's modulus / MPa	Failure strength / MPa
Dry / unloaded	0	0	1851	46.8
Dry / loaded	0	46	1723	46.3
Wet / unloaded	2.95	0	1595	38.9
Wet / loaded	3.75	46	1419	36.4

4. Finite element modelling

The numerical modelling procedure in this work was performed in two steps: 1) to model

the long-term ageing process in the adhesively bonded joints under combined thermal-hygro-mechanical service loading conditions using a fully-coupled methodology; 2) to simulate the quasi-static tensile loading process in adhesive joints (aged in Step 1) using a CZM model. Modelling details related to Step 1 can be found elsewhere [32] and this paper focuses on the modelling procedure performed in Step 2. However, key data from Step 1 will be presented.

4.1 The SLJ model

The 3D finite element (FE) model was built in the FE package Abaqus® to simulate both steps outlined in the paragraph above. It is necessary to have two models (one for each Step) as the adhesive elements are different (continuum elements in Step 1 and cohesive elements in Step 2). However, it is necessary to transfer results from the Step 1 model into the Step 2 model. To achieve this transfer of data it is important to provide the same node numbering sequence. The numerical analysis was carried out assuming geometric non-linearity. Fig. 3 shows a typical FE mesh for the SLJ with refinement around the bonding areas where large peel and shear stress gradients are located [27]. Exactly the same mesh refinement was used in the models for both Steps (the only difference was that the adhesive used a different element type in each model). Reduced linear 3D stress elements (C3D8R) were employed for the substrates and, in Step 2, 3D cohesive elements (COH3D8) were used for the adhesive to include the bilinear CZM. It is worth noting that a higher mesh density (0.20 mm×0.20 mm×0.17 mm) was utilised for the elements around the bonding area to provide more accurate results.

For the Step 2 analysis one end of the substrate was restrained with an encastre constraint, while the other end was constrained with a kinematic coupling to allow only the translational displacement in the x direction for the nodes at this end. A displacement boundary condition was applied at the control point of the kinematic coupling to simulate the quasi-static loading process in the Instron universal testing machine. This displacement was ramped until the joint completely failed. In the Step 1 analysis a constant (creep) load was applied instead of the ramped displacement.

Fig. 3. FE mesh for the single lap joint showing detailed refinement around the bonding area.

4.2 Cohesive zone model

The cohesive zone model was first introduced by Barenblatt [33-34] and incorporated into a computational framework by Hillerborg et al. [35]. Incorporating a small process zone ahead of the crack tip where material yielding, micro-cracking, and void formation is observed, the CZM is frequently used in numerical analysis. In this work, due to the substrate surface pre-treatment method introduced in Section 2.1, failure in both unaged and aged SLJs was primarily cohesive in the adhesive, as can be seen from the fracture surfaces shown in Fig. 4. It can be seen that adhesive remained evenly on both of the substrates and fracture generally occurred within the adhesive layer. Thus, in the residual strength FE analyses the adhesive layer was modelled with a cohesive elements that adopted a bilinear traction-separation response, as shown in Fig. 5a. This consists of three main parameters: the elastic stiffness K , the tripping traction T_{max} and the fracture energy G_C . This approach could be adapted in further modelling work to include interface failure modelled using a very thin cohesive elements with the adhesive modelled using solid elements that adopt a continuum damage constitutive model. These approaches have already been used separately for interfacial and cohesive failure [26, 18] respectively.

Fig. 4. The fracture surfaces of (a) unaged and (b) aged SLJs after quasi-static tensile testing.

The bilinear traction-separation law provides an initial linear elastic behaviour followed by a linear damage evolution. Damage initiation can be specified through different criteria. In this work, the quadratic stress criterion was introduced as expressed in Eq. 3.

$$\left\{ \frac{\langle T_n \rangle}{T_{n-\max}} \right\}^2 + \left\{ \frac{T_s}{T_{s-\max}} \right\}^2 + \left\{ \frac{T_t}{T_{t-\max}} \right\}^2 = 1 \quad (3)$$

Here, subscripts n , s and t indicate the normal, first and second shear directions of the traction (T) respectively, and $\langle \rangle$ is the Macaulay bracket meaning that a compressive stress does not lead to damage initiation. After initiation occurs, a linear softening stage follows. Complete separation of the adhesive layer is determined by a quadratic power-law criterion as defined in Eq. 4.

$$\left(\frac{G_I}{G_{IC}}\right)^2 + \left(\frac{G_{II}}{G_{IIC}}\right)^2 = 1 \quad (4)$$

here, G_I and G_{II} are the energies released by the traction for mode I and mode II loading respectively.

4.3 Determination of the degraded CZM parameters

A degraded bilinear traction-separation CZM, as shown in Fig. 5, was used to characterise the debonding process in the aged SLJs. The CZM was assumed to be degraded by moisture ingress (Fig. 5b) and the degraded CZM properties were then further reduced to accommodate the creep damage caused by the sustained loading (Fig. 5c).

The calibration process of the CZM parameters for the unaged adhesive has been reported elsewhere based on the experimental data from unaged bulk adhesive tensile tests and the double cantilever beam test [23]. The degraded normal elastic stiffness (K_n) was obtained through the degraded Young's modulus (E) in the aged bulk adhesive (as shown in Table 6) divided by the cohesive element thickness. Similarly, the aged first and second shear elastic stiffness (K_s and K_t) were calculated through the aged shear modulus G (determined from the Young's modulus E) divided by the cohesive element thickness. The decrease in the tripping tractions (T_{n-max} , T_{s-max} and T_{t-max}) and the fracture energies (G_I and G_{II}) were assumed to be linearly proportional to the drop in the static failure strength of the aged bulk adhesive [17, 34-35].

Fig. 5. Schematic of the degraded bilinear traction-separation CZM.

Table 6 CZM parameters for the unaged and aged FM73 adhesive.

Degradation environment	K_n N/mm ³	$K_f=K_s$ N/mm ³	T_n MPa	$T_f=T_s$ MPa	G_{IC} kJ/mm ²	$G_{IIC}=G_{IIC}$ kJ/mm ²
Dry/unloaded	10000.0	3575.0	63.5	36.3	2.50	5.00
Dry/loaded	9304.6	3326.4	62.9	35.9	2.47	4.95
Wet/unloaded	8613.1	3079.2	52.8	30.2	2.08	4.16

Wet/loaded	7664.2	2740.0	49.5	28.3	1.95	3.89
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4.4 Importing the degradation data from the ageing phase (Step 1)

Before performing the quasi-static test, the aluminium SLJ was immersed in deionised water at 50 °C and loaded at two constant loading levels (12% and 17.5% of the static failure load) for 6 months (Step 1). It is worth noting that although the stress states in the bulk adhesive and adhesive layer in the joint are different, as a starting point the von Mises stress was used to characterise the stress dependence of the moisture uptake. This procedure was carried out in Abaqus and has been discussed in detail elsewhere [32].

In order to evaluate the degradation effect induced by the hostile environment, the accumulated adhesive degradation data from the Step 1 model should be considered and imported into the Step 2 model as field variables for the adhesive. A number of researchers have focused on the decrease in the mechanical properties of adhesive structures caused by moisture ingress and external loading [11, 16, 38]. In this work, the accumulated a) moisture uptake and b) creep strain in the adhesive layer were selected as the degradation factors (DFs) in the Step 2 model.

The moisture uptake and equivalent creep strain were defined as field variables (FV2 and FV4 respectively) in the Step 1 model and output to a results file with the values averaged at the nodes of the adhesive layer. In order to import this data into the Step 2 model correctly, exactly the same node numbering sequence between the Step 1 and Step 2 model was required. In Step 2 this was achieved by inheriting the FE model from Step 1 but modifying the adhesive element type from a 3D continuum to a 3D cohesive element.

With the results file obtained from Step 1, the relevant degradation data were selected and output into two separate .FIL files (moisture uptake and creep strain data respectively) with a Fortran user subroutine ABQMAIN in conjunction with the Abaqus command function. These two .FIL files were then imported directly into the Step 2 model as predefined field variables, defining the moisture uptake and equivalent creep strain distribution in the adhesive layer inherited from the degradation process. The contours showing the distribution of the moisture content and equivalent creep strain in the adhesive layer at the end of Step 1 and the beginning of Step 2 models after 6 months immersion and simultaneous loading at 12% of the static joint strength are presented in Fig. 6. It can be seen that the original and imported contour distribution for both DFs were in good agreement with each other with minor difference on the edges of the adhesive layer, thus lending confidence to the data transfer process. It should be noted that the moisture content in Fig. 6a is reasonably uniform across the adhesive layer (consider the small range on the contour values) as might be expected after such a long period of exposure with a reasonably thin (widthwise) joint. Further it should be noted that Fig. 6a plots the actual moisture mass content (in %) rather than the more usual normalised moisture concentration. These results are discussed in a little more detail in Section 5.1.

Fig. 6. The distributions of (a) the moisture uptake and (b) equivalent creep strain in Step 1 and Step 2 models after 6-month ageing.

4.5 Concept framework of the FE modelling

A general framework illustrating the modelling techniques utilised in Step 2 is shown in Fig. 7. It can be seen that the degraded i) elastic stiffness K , ii) tripping traction T and iii) fracture energy G_c , (which constitute a set of CZM parameters) are dependent on the prior ageing (moisture and loading level) experienced. Using the data transfer procedure outlined in Section 4.4, the DF (moisture uptake and equivalent creep strain) distribution in the aged adhesive layer was determined and transferred from the validated simulation in the Step 1 model [32] into the Step 2 model. Finally the quasi-static tensile testing of the degraded aluminium SLJ was simulated numerically using progressive damage FE modelling to obtain the predicted residual strength after ageing.

Fig. 7. Concept framework of the FE modelling (DF being the degradation factor).

5. FE modelling results and discussion

The SLJ simulation in Step 1 (the ageing stage) provided reasonable moisture and stress distributions and good correlation with the experimentally measured joint creep extension-time curve. Detailed verification and discussion of the modelling technique have been reported elsewhere [32], however major modelling achievements in Step 1 are presented in Section 5.1 to provide a better understanding of the whole modelling work. Following this, the majority of this section focuses on the SLJ simulation results from Step 2 (the residual strength modelling) to investigate the degradation in joint strength induced by the prior simultaneous exposure to moisture and sustained loading.

5.1 Major modelling achievements in the ageing phase (Step 1)

The distribution of the normalised moisture concentration (c) along a path through the centre of the adhesive layer in the widthwise direction for the joint loaded at 12% of the static failure strength is shown in Fig. 8a. It can be seen that no moisture exists in the centre of the adhesive initially, while after 12 and 42 days water diffuses into the adhesive with c in the centre reaching around 0.4 and 0.8 respectively and the adhesive layer reaching full saturation after 3 months exposure.

The distributions of the actual mass uptake of moisture across the overlap width in both unloaded and loaded (12% of the static failure load) conditions are shown in Fig. 8b. The moisture diffusion rate is reduced in the unloaded joints due to the absence of external loading. However, the reduction in the moisture content level is not as much as might be expected from the saturation contents in Table 3. This is due to the fact that, even in the absence of any external loading, internal stresses are induced from the simultaneous thermal and swelling expansion process in the adhesive layer, which leads to an increase in moisture diffusion rate and saturation value in the mechanically “unloaded” joints.

The von Mises stress distributions along a path in the centre of the adhesive layer in the lengthwise direction after 0 days (initial), 12 days and 42 days for the joint loaded at 12% of the static failure strength is presented in Fig. 8c. It can be seen that the stress concentration observed initially at the end of the bonding area reversed after 12 days exposure and then largely reduced after 3 months environmental degradation. This can be explained by the high creep rate in the highly stressed and immersed region at the overlap ends. The subsequent smoothing of the von Mises stresses can be explained by a combination of the same mechanism and the fact that as the moisture diffuses further into the joint there is less differential swelling along the adhesive layer and hence a more uniform transverse (peel) adhesive stress. After 3 months the adhesive layer is essentially saturated and the stress is nearly uniform, thus the joint continues to creep at a uniform rate across the entire adhesive layer.

The experimental and predicted creep responses in the SLJs under different loading levels (12% and 17.5% of the static failure strength) are presented in Fig. 8d. With the creep

parameters shown in Table 4, reasonable agreement is observed between the simulated and experimentally-measured curves over the degradation period, particularly at the higher loading level.

Fig. 8. The distributions of (a) the normalised moisture concentration, (b) the moisture uptake, (c) the von Mises stress in the adhesive layer and (d) the experimental and predicted creep responses in the SLJs.

5.2 Strength degradation

The predicted load-displacement curves and the comparison of the experimental and predicted peak load for the unaged and 6-month aged SLJs are presented in Figs. 9a and 9b respectively. It can be seen in Fig. 9a that moisture uptake on its own caused significant degradation in the peak load of the aged SLJ (a reduction of 18.5%) as well as the displacement when the peak load was achieved. However, although the applied constant loading (either 12% or 17.5% of the failure loading levels) did provide further degradation the predicted joint residual strength was not lowered significantly. This can be clarified in Fig. 9b where the 6-month immersion at 17.5% of the failure loading only slightly further decreased the joint static strength from the 12% set in both predicted and experimental results (only leading to a further 0.8% and 2.6% reduction respectively). It can also be seen in Fig. 9b that although the predicted peak load was slightly higher than the experimental one for all conditions, generally good agreement was achieved between the predicted and experimental SLJ residual strengths for unaged, wet-12% failure load and wet-17.5% failure load ageing

conditions (with the differences being 0.20%, 3.65% and 6.26% respectively).

Fig. 9. (a) The predicted load-displacement curves and (b) the comparison of the experimental and predicted peak load for the unaged and 6-month aged SLJs.

5.3 Stress and damage distribution

Fig. 10 shows the distribution of (a) the von Mises stress and (b) the cohesive zone damage (SDEG) on the adhesive mid-plane along the overlap length at the peak load for the unaged and aged (wet-unloaded and wet-12% failure load) SLJs. It should be noted here that damage refers to the damage caused by the static load. The environmental damage has already been applied by scaling down the CZM parameters. It can be observed from Fig. 10b that, for all conditions, the damage in the adhesive layer started at the overlap end and progressed towards the centre of the bonding area with the SDEG value higher at the edge than the middle region. This difference was more obvious for the aged than the unaged SLJ, which was due to the fact that for the aged SLJs the environmental reduction in the CZM properties was always higher at the edge than the middle. It can also be seen from Fig. 10b that a fully damaged state (SDEG=1) is not reached along the overlap length at the point of peak load. This may be because any further damage reduces the overall stress level in the adhesive layer resulting in a further damage evolution but at a lower load.

Comparing Figs. 10a and 10b, it can be seen that the von Mises stress was lower at the overlap end. This is because the damage is highest at the overlap ends and this degrades the material strength (see the traction separation response, Fig. 5). For the aged conditions, which (at peak load) have not damaged across the entire overlap length, it is also worth noting that the von Mises stress reached its peak value at the damage tip. This is consistent with the trend reported in other research [15]. However, for the unaged condition, the von Mises stress kept increasing towards the middle of the adhesive layer as no zero-damage zone was encountered.

Fig. 10. The distributions of (a) von Mises stress and (b) the static damage (SDEG) along the overlap length at the peak load for unaged and 6-month aged SLJs.

5.4 Evolution of stress and damage in the aged SLJ

Fig. 11a shows the contours of the von Mises stress and the static damage in the adhesive layer for the aged (wet-12% failure load) SLJ at point 2 (the peak load) in the load-displacement curve shown in Fig. 9a. It can be seen from Fig. 11a that both stress and damage were symmetric in the overlap width direction and varied gradually lengthwise. Figs. 11b and 11c show the von Mises stress and the damage for the same joint (wet-12% failure load) along the centre line of the adhesive in the lengthwise direction for points 1, 2 and 3 on Fig. 9a. At point 2 (the peak load) damage was already found at the overlap end while the centre area remained undamaged (Figs. 11a and 11c). The von Mises stress (Fig. 11b) kept increasing during the quasi-static loading process until it reached the peak value at Point 2 (the peak load) and then decreased as the damage propagated towards the centre region. The damage propagation process in the adhesive layer can be seen in Fig. 11c. The damage remained zero across the adhesive layer at point 1. By the time the peak load was reached the damage at the overlap edge had an approximate value of 0.5. Although the damage zone length reached around 3.5 mm inside the overlap, the centre area remained undamaged at point 2 (the peak load). Then, with further joint extension, the damage increased and propagated towards the centre of the adhesive layer and led to a globally-high damage level (around 0.9) across the overlap region at point 3. At this level of damage the traction that can be sustained (and thus the load carried by the joint) has been reduced considerably. Finally complete failure was achieved almost simultaneously across the whole bonding area with the damage value reaching 1.0.

Fig. 11. The contours of (a) von Mises stress and the static damage at point 2 and (b) the distribution of von Mises stress and (c) the static damage along the overlap length in the aged SLJs at the selected points shown in Fig. 9a.

5.5 Evolution of stress and damage in the unaged SLJ

Figs. 12a and 12b show the von Mises stress and the damage for the unaged joint along the centre line of the adhesive in the lengthwise direction for points 1, 4 and 5 on Fig. 9a. It can be seen comparing Figs. 11b and 12a that the stress concentration at the overlap edge was more severe in the unaged SLJs than the aged ones at point 1 and that at the peak load the stress levels are higher in the unaged joint. This is because the initial stiffness and the tripping tractions were higher in the unaged adhesive layer according to the values shown in Table 6. The damage evolution in the unaged SLJs (Fig. 12b) was quite similar to the aged one (Fig. 11c), the main difference being that at the point of peak load (point 4) damage existed through the whole adhesive layer with a minimum SDEG value around 0.5. This is because in the unaged joint there is no moisture to preferentially weaken the region at the end of the overlap, so localised damage is less likely.

Fig. 12. (a) The distribution of von Mises stress and (b) the static damage along the overlap length in the unaged SLJs at the selected points shown in Fig. 9a.

6. Conclusions

Experimental and numerical modelling work has been carried out to investigate the

strength degradation of adhesively bonded joints in a combined thermal-hygro-mechanical service condition. To the authors' best knowledge this is the first case of fully-coupled environmental degradation modelled for adhesively bonded joints.

The experiments performed on bulk adhesive specimens provided the necessary material parameter dependency on the environmental factors, while the tests on SLJs were utilised as a validation of the numerical results. The numerical modelling was performed in two steps: 1) to simulate the ageing environment and the consequent environmental degradation process; 2) to simulate the strength reduction caused by the long-term ageing, i.e. the residual strength. This paper focused on the second step and a reliable importing technique was utilised to transfer the selected environmental degradation data from Step 1 into Step 2. The quasi-static tensile testing on the unaged and aged SLJs was successfully modelled with a degraded CZM in which the cohesive parameters were dependent on the local moisture uptake and creep strain distributions in the adhesive layer (Tables 5 and 6). The experimental results (Fig. 8) show that the existence of water significantly degraded the joint strength while the introduction of sustained load further extended this effect, but to a less significant extent (Fig. 8b). Good correlation was found for the predicted and experimental results.

Modelling the joint failure was based on a cohesive failure mode in the adhesive layer. The modelling technique adopted in this current work can be extended to simulate interfacial failure and even mixed failure modes, as outlined in Section 4.2. Another simplification that can be further investigated is using the von Mises stress to control the stress dependence of the moisture diffusion. The bulk adhesive and the SLJ have a different stress states and thus other stress measures might yield a different response. Future work can also be focused on extending the developed experimental and modelling technique onto aged joints with longer and wider overlap lengths to investigate the strength degradation of SLJs with partial saturation and local failure rather than the global failure mode that occurred in this study. Also, strength reduction of SLJs under simultaneous fatigue loading in a wet environment has always been an important subject in the durability of adhesive joints and forms a natural extension to this work.

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