Microdamage analysis in thermally aged CF/polyimide laminates

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Abstract. Microdamage in layers of CF Thornel® T650 8-harness satin woven composite with thermosetting polyimide NEXIMID® MHT-R resin was analysed. After cooling to room temperature multiple intra-bundle cracking due to tensile transverse thermal stresses was observed in the studied [(+45/-45)/(90/0)]2s composite. The composite was subjected to thermal cycling quantifying the increase of crack density in layers. Comparison of two ramps with the same lowest temperature shows that the highest temperature in the cycle has a significant detrimental effect. Exposure for 40 days to 288°C caused many new cracks after cooling down to room temperature. Both aged and not aged specimens were tested in uniaxial quasi-static tension. Cracking was analysed using fracture mechanics and probabilistic approaches. Cracking in off-axis layers was predicted based on Weibull analysis of the 90-layer. The thermal treatment degraded the cracking resistance of the surface layer and of the next layer.

1. Introduction

An increasing interest of aerospace industry in composite materials for applications in harsh environments has been driving improvement and development of new composites. Composites designated for aero-engine applications considered in the current paper are subjected to high temperatures. Long exposure to elevated temperatures can cause irreversible changes in the morphology (termed aging in this paper) reducing strength and stiffness [1]. In addition, cyclic variations between high and low temperatures can lead to fatigue resulting in microdamage accumulation [2].

The effect of thermal aging on mechanical performance of CF/RTM6 resin and CF/BMI resin cross-ply laminates was studied in [3] showing that the intralaminar crack density in a following tensile test is a clear function of aging time. The elevated temperature level during thermal cycling with a fixed lowest temperature has an accelerating effect on the microdamage development [4] which currently does not have solid explanation. Apparently irreversible phenomena take place at the highest temperature and microdamage evolves when the lowest temperature in the cycle is reached.

The main objective of the current work is to study intralaminar cracking during thermal cycling and tensile quasi-static loading of “as received” and “aged” quasi-isotropic carbon fibre/polyimide woven composites. Microcracking initiation and growth will be analysed using Weibull initiation strength distribution approach and fracture mechanics, focussing on

- Differences in crack density growth in different layers
Analysing the effect of high temperature exposure on intralaminar cracking

2. Experiments

2.1. Description of materials.
Quasi-isotropic \([+45/-45]/(90/0)\)\(_2\) composite was manufactured (RTM) using 8-harness satin Cytec T650 CF and thermosetting polyimide resin denoted NEXIMID® MHT-R (MHT-R). The material was cured at 340°C for 30min followed by 2.5h post cure at 370°C using 12 bar pressure. In Figures the composite is labelled as 432 plate. The volume fraction of fibres is around 60%. More details on manufacturing and properties of the laminate are given in [5]. The average thickness of one “layer” (bundle) is approximately 0.18 mm. The bundle width is about 1.5mm and the aspect ratio partially justifies the used “layer assumption”. Both edges of all specimens had fine polishing for damage quantification using optical microscopy. Part of specimens was subjected to aging at 288°C for 960 hours (40 days) in air environment, afterwards cooled down to room temperature (RT) for inspection.

2.2. Experimental procedures.
Two different thermal cycling ramps were used. In the ramp (R1) the temperature was varied between -60°C and RT whereas in ramp (R2) the change was between -60°C and 288°C. The heating and cooling rate was approximately 100°C for minute and the specimen was exposed to each constant temperature for 10 minutes for up to 150 cycles.

Tensile loading and unloading tests with increasing maximum were performed at RT with displacement rate 2mm/min. Strains were measured using extensometer with gauge length 50mm. The damage state in all specimens was analyzed before any treatment (notation not aged (NA)) and also after aging (A). After certain number of cycles in thermal fatigue and after each loading-unloading step in quasi-static tensile loading the specimen was inspected using optical microscope. The respective number of cracks \(N_i\) over distance \(L\) (50mm) on the specimen edge was counted and the crack density in each layer was calculated as

\[
\rho_i = \frac{N_i}{L \sin \theta_i}
\]

where \(\theta_i\) is the fiber orientation angle in the layer. The void content was measured using image analysis software (NIS-elements BR) together with optical microscope. The void content was defined as the area fraction of voids on the observed edge surface. The average void content was measured over the area of about 100mm\(^2\). From one specimen to another, the average void content varied from 0.3% to 2.4% which is a very high variability.

3. Results and discussion

3.1. Initial damage state.
The initial damage state is caused by thermal stresses at the lowest temperature. According to the Classical Laminate Theory (CLT) thermal transverse stresses in a quasi-isotropic laminate is the same in all layers and the in-plane shear stress is zero. Hence, the number of initiated cracks statistically should be the same in all layers. Still, on the specimen edge the crack density in different layers was different: much higher crack density in the surface +45-layers; lower in the 90-layers but still larger than in the internal \(\pm 45\)-layers, where it was very low. To explain these observations, 3-D FEM thermal stress analysis was performed to study free edge effect on thermal stress distribution: \([+45/-45]/(90/0)\)\(_2\) specimen was modelled (a specimen cut from the central part of the plate has free edges where the stress state is different than in the same position in the plate). Clear differences between transverse edge stresses in \(\pm 45\)-layers and in 90-layers were found, see details in [6]. In 90-layers the transverse stress is magnified by approximately 20% whereas in all 45-layers it is only 30% of the CLT stress. This means that in the specimen new cracks may have been created in the edge regions of
the 90-layer. In 45-layers new cracks were not introduced by making specimens. Therefore, the crack density in the central part of the 90-layer should be similar to the damage state observed on the edge of the 45-layer.

To explain the much higher crack density in the surface layers we have to distinguish two phases in crack evolution: initiation (mainly transverse growth) and propagation along fibers. The propagation of a crack may be described by Linear Elastic fracture mechanics (LEFM) which states that the crack will propagate when the energy release rate (ERR) $G$ reaches or exceeds the critical value $G_C$. The crack initiation process is stochastic and characterized by coalescence of fibre/matrix debonds leading to “defect” creation and growth in the ply thickness direction. According to LEFM the stress required for central crack growth in an infinite plate is $1.12\sqrt{2}$ times higher than for an edge crack of the same size in a semi-infinite plate [6]. Generalizing this result to laminates we can expect that under thermal loading more cracks are initiated in the surface layer of the quasi-isotropic laminate. The available energy for crack propagation along fibres depends on the ply thickness and on the position of the layer: in the surface layer it is about 2.4 times larger [7]. Hence, the propagation of an initiated crack in the surface layer would start at much lower transverse stresses.

3.2. Thermal fatigue.

Each data point for crack density in Fig. 1 corresponds to the average on two edges of one specimen for all layers of a given orientation. For all layer orientations the R2 (-60°C to 288°C) ramp induced more damage than R1 (-60°C to RT). This means that the temperature interval, $\Delta T = T_{\text{min}} - T_{\text{max}}$ at fixed $T_{\text{min}}$ has effect on the cracking in thermal fatigue. It is consistent with the Paris law for growth of individual crack: $\Delta G$ is larger for R2 ramp than for R1 ramp. The crack density in thermal cycling was the highest in surface layers followed by 90- and even lower in internal 45-layers (not shown). Much larger crack density in surface layers was observed also in [8] where thermal cycling at cryogenic temperatures was performed.

In fatigue, certain number of cycles $N_{\text{init}}$ is required to initiate a crack and a different number of cycles $N_{\text{prop}}$ is required for its propagation along fibres. If $N_{\text{init}} \gg N_{\text{prop}}$, the number of cycles for full development of the crack is initiation governed. The initiation in the surface layer is accelerated by the free surface effect. The ERR for crack growth along fibres is also much higher in the surface layer than in internal layers and initiated cracks almost instantly grow through the specimen. To investigate whether the edge cracks propagate inside the specimen and vice versa, the edge of one specimen subjected to 150 cycles in R2 was polished to quantify the damage state versus the distance $d$ from the specimen edge: a) the crack density inside the specimen was approximately the same in the 90- and internal 45-layers; b) the crack density on the edge of the surface 45-layer does not change with the distance from the edge, proving that in this layer there is an excess of ERR for propagation.

3.3. Enhanced temperature.

Specimens were exposed to 288°C in ambient air environment (aged) and cooled down to RT to count cracks on specimen edges, see Fig. 2. The thermal stresses at RT introduced new cracks. Surprisingly the crack density on the edge of the 90-layer is higher than in the surface +45-layer reaching about 1.7cr/mm. Edge polishing of a 40 days aged specimen was performed to establish whether the edge data are representative for the bulk of the composite. The results show that in the 90-layer the amount of cracks away from edge is very similar to the number of cracks found on the edge of the same specimen before aging. It indicates that in the bulk of the 90-layer the material degradation (resistance to cracking at room temperature) due to high temperature exposure for 40 days is rather small: cracks initiated on the degraded edge do not propagate inside the layer.

There was no edge “concentration” in crack density for surface layers. During the aging test the whole surface 45-layers are in contact with the environment. Therefore more uniform material degradation can be expected in surface layers leading to more uniform resistance to crack initiation and propagation.
Figure 1. Edge crack density growth in two thermal fatigue ramps: R1 (-60;RT)°C; R2 (-60;+288)°C.

The degraded zone propagates in thickness direction from the specimen surface and the crack density in internal 45-layer depends on the distance of this layer from the surface (the crack density in the 45-layer next to surface layer is much higher than in 45-layers close to the symmetry plane of the laminate where the crack density is very similar as in 90-layers). This means that not only the surface layer but also the next layer has degraded, which is expected because the thickness of one layer (0.18mm) is much smaller than the size of the observed highly degraded zone at the edge (about 1 mm).

Figure 2. Crack density on the specimen edge versus aging time: a) surface 45-layers; b) 90-layers.

3.4. Damage state in tensile mechanical loading.

The severely damaged edge zone of the aged specimens was removed by polishing before mechanical testing. The effect of aging on cracking resistance was expected to be high in surface layers. The tensile loading introduced new cracks on the edge of aged (A) and not aged (NA) specimens. The cracking data presented below are based on edge observations only. To check that edge data are representative the damage statistics on the edge was compared with the damage state (after polishing) at several distances from the edge finding that statistically the crack density is not reducing.
Apparently most of the initiated cracks had enough of ERR to propagate along the fibre direction and that crack initiation should be the focus for the used composite. These results give sufficient confidence in that edge data are representative for the damage state inside the specimen.

Thermal stresses do not change during the tensile test whereas the mechanical stress is proportional to the applied strain. According to CLT at 1% strain the total transverse stress/shear stress ratio in the 45-layer is \( \sigma_{LT}^{45}/\sigma_T^{45} \approx 0.45 \) and at lower strain the fraction of the shear stress is lower. For this reason, analysing failure in layers we will neglect the shear stress contribution.

The conditions for crack initiation in surface and internal layers are different: in internal layer the surface defect growth is suppressed by constraint from the adjacent layers and defects in the middle of the layer will grow into a crack. In [9] relationship between crack initiation stress in internal and surface layers was suggested

\[
\sigma_{T,init}^{internal} = k \sigma_{T,init}^{surface}
\]

where \( k = 1.12\sqrt{2} \) for surface layer and \( k = 1 \) for internal layer. The initiation of a crack is a stochastic process with certain probability and with random position. In simulations of crack initiation we assume that any layer in its transverse direction can be considered as consisting of a chain of elements following the two-parameter Weibull strength distribution

\[
P_f = 1 - \exp \left( -\left( \frac{k \sigma_T}{\sigma_{ino}} \right)^m \right)
\]

Parameter \( k \) is introduced in (3) to reflect the favourable situation for crack initiation in the surface layer. If the crack density in layer is low enough to allow for large plateau regions in stress distribution, the probability of failure at transverse stress \( \sigma_T \) can be estimated using expression [8]

\[
P_f = \frac{\rho(\sigma_T)}{\rho_{max}}
\]

In (4) \( \rho_{max} \) is the assumed maximum possible crack density in a layer when due to interaction the transverse stress between two neighbouring cracks is too low to lead to new intralaminar crack formation. Based on experimental observations \( \rho_{max} \approx 1/t \), where \( t \) is the thickness of the layer. Using the experimental crack density dependence on transverse stress to calculate \( P_f \), we plot the \( \log(-\log(1-P_f)) \) versus \( \log(\sigma_T) \) and use the standard procedure to estimate Weibull parameters. The result is rather sensitive to the used data points on both extremes: data points at high crack density are affected by stress perturbations from neighbouring cracks while the stress at first crack varies a lot from specimen to specimen. The estimates of Weibull parameters can be used in more accurate Monte-Carlo simulations over the whole crack density region including the high crack density region where the stress perturbations caused by cracks are overlapping.

Cracking was analysed in 90-layers converting the strain dependence of crack density to transverse stress dependence using CLT and the estimated thermal stresses. Using (4) the crack density data was transformed to probability of failure and (3) was used to estimate Weibull parameters (referred as \( P_f \)-approach). Weibull parameters for both aged and not aged composite are given in Table 1.

3.4.1. Damage accumulation analysis starts with plotting the log-log relationship for 90-layers in NA specimens as shown in Fig. 3a. The first point on the left corresponds to crack density caused by thermal stresses which has rather uncertain value. The last three points deviate from linearity which is a sign of crack interaction. According to Fig. 3b, where the test data and the simulation using (4) are shown, deviation starts at \( \rho_{00} \approx 1.6 \text{ cf/mm} \). In general the obtained Weibull parameters together with (3), (4) give good description of the cracking development.

The obtained Weibull parameters were used to predict crack density in internal +45-layers and in the surface +45-layer of the NA laminate. All internal +45-layers behave in a very similar way and the
presented data are averages. The data and simulations using the \( P_f \)-approach are presented in Fig. 4 which shows good agreement proving that cracking in +45-layers has the same transverse stress dependence as in 90-layers. The crack density in the surface +45-layer, shown in Fig. 4b is significantly higher. The \( P_f \)-approach with \( k = 1 \) gives too low crack density. An attempt to use in simulations equation (3) with \( k = 1.12\sqrt{2} \) failed – far too high crack density was predicted, see Fig. 4b. These results show that the role of the free surface on defect growth has been oversimplified in (2).

Table 1 Weibull parameters from 90-layer cracking

| Composite | \( m \) | \( \sigma_{in0} (\text{MPa}) \) |
|-----------|-------|-----------------|
| 432-NA    | 7.84  | 135.2           |
| 432-A     | 6.88  | 134.0           |

Figure 3. Cracking in 90-layers (NA-432): a) Weibull plot of test data and linear fit; b) crack density versus transverse stress \( \sigma_{T90} \), \( P_f \)-approach (4).

3.4.2. Effect of aging on damage development in tension was studied obtaining Weibull parameters for 432-A specimens, see Table 1, from aged 90-layers cracking data. They were used in the \( P_f \)-approach to predict crack density growth in 45-layers, see Fig. 5. For internal +45-layers predictions are in good agreement with test data. Similarly as for NA specimens, the crack density in the surface layers could not be predicted: it is even outside the previously discussed bounds. The crack density in these layers a) is much higher than in the surface layers of an unaged specimen and b) the difference in crack density with internal layers of the same orientation is much larger than before the aging test. That means that the surface layer has different failure properties than the internal layers: it has lower initiation stress and therefore \( \rho \) is much higher than in NA case. The probability of transverse failure in 90-layers of not aged and aged composite is very similar: the high temperature exposure has not affected the damage behaviour of 90-layers.

In Fig. 6 crack density is shown separately in the L3 and in the L7-layer, both with \(-45^\circ\) orientation. The L3 layer is the third layer from the midplane of the \([+45/-45/90/0/+45/-45/90/0]\), laminate and L7 is the seventh layer from the midplane. Thus, the L7 is close to the surface. The \( P_f \)-approach simulation describing cracking in +45-layers, is shown as solid line. For not aged specimens the crack density in L3 layer is just slightly lower than in the L7 layer. In aged specimens in Fig. 6b the
The difference is striking: much higher crack density in L7 layers. In the L3 layers the cracking is similar as for not aged -45-layers. An obvious explanation for this is aging: layers close to the laminate mid-plane have not degraded. Only the surface layer and the following L7 -45-layer are aged. There are more cracks in -45-layers than in +45-layers which we explain by the effect of damage in adjacent 90-layers.

**Figure 4.** Crack density versus strain in 45-layers of NA-432: data and predictions using Pf-approach.

**Figure 5.** Microcracking in aged A-432 composite.

### 4. Conclusions
Microscopy was used to analyze the damage state in quasi-isotropic CF/polyimide 8-harness satin composite after manufacturing and its evolution a) due to exposure to 288°C for 40days; b) due to thermal cycling; c) in result of tensile mechanical loading. In all cases much higher rate of crack density growth was found in surface layers because of the free surface effect on initiation and also on the propagation of intralaminar cracks. Due to temperature exposure about 1mm of the edge zone was severely degraded causing more cracks whereas damage inside the specimen was not affected.
In tensile loading the crack density in different layers of the laminate was analyzed using for crack initiation Weibull transverse strength distribution. It was found that cracking in internal +45-layers can be predicted based on Weibull properties of 90-layers. The crack density in the surface +45-layer was much higher even for not aged specimens. The thermal treatment significantly reduced the resistance of the surface layer to cracking under mechanical tension. It degraded also the layer next to it, whereas failure resistance of central layers was not affected.

![Cracking in -45-layers of the 432 quasi-isotropic laminate (layers are numbered L3, L7 regarding their position with respect to mid-plane): a) NA; b) A.](image)

**Figure 6.** Cracking in -45-layers of the 432 quasi-isotropic laminate (layers are numbered L3, L7 regarding their position with respect to mid-plane): a) NA; b) A.

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