THE EFFECT OF HIGH TEMPERATURE ON THE MECHANICAL PERFORMANCE OF NOVEL HIGH T_g POLYIMIDE-BASED CARBON FIBRE-REINFORCED LAMINATES

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Abstract

In this study, the outcomes from the mechanical testing of the carbon fibre-reinforced polyimide composite system T650/NEXIMID® MHT-R at ambient and elevated temperatures are presented. These results are compared to assess the effect of mechanical loading at 320°C on the performance of the system in tension, compression and Short-Beam Shear. The experimental campaign indicated that the mechanical loading at 320°C had a trivial effect on the tensile properties (fibre-dominated) whilst a more pronounced effect was noted on the compression and Short-Beam Shear (matrix and fibre/matrix interface-dominated properties).

1. Introduction

Most organic polymers used nowadays extensively in the aeronautics industry for high performance fibre-reinforced composites are limited to temperatures in the area of 120°C. Beyond that temperature their mechanical performance is significantly deteriorated due to softening while at even higher temperatures they start to decompose compromising thus the overall integrity of the composite structure [1,2]. For this reason polymer-matrix composites have been limited to structural components with temperature requirements well below that temperature zone, rendering them as unsuitable candidates for applications with higher temperature requirements such as turbine engines. Nevertheless, polymers with glass transition temperatures in the excess of 250°C (cyanate esters and bismaleimides) or even over 400°C (phthalonitriles and polyimides) have been available for several years [3], however issues relating to processing and low toughness have hindered them from being utilised in structural applications exposed to high temperatures [3,4].

In the recently completed project HicTac (High performance composites for demanding high Temperature applications) funded by CleanSky [5], carbon fibre-reinforced/polyimide composites (T650/MHT-R) exhibiting a T_g in the range of 370-400°C were developed whilst the outcomes have been disseminated [6-8]. The current communication follows up on a previous study by the authors [6] on the initial assessment of the mechanical performance of the aforementioned T650/MHT-R carbon fibre-reinforced/polyimide composites and the effect of the epoxy-based sizing on failure process. In this study, quasi-isotropic T650/MHT-R laminates were manufactured using a modified curing routine to that previously reported by the authors [6,7]. Subsequently the laminates were tested in tension, compression and Short-Beam Shear at ambient temperature and at 320° in order to evaluate the effect

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of exposure to high temperatures on the mechanical performance of these polyimide-based composites. In addition, fractographic analysis of compression and Short-Beam Shear specimens was carried out to provide information about the dominant failure mechanisms and effect of high temperature on the failure process.

2. Experimental

In this chapter, a description of the material systems and manufacturing process as well as the experimental procedures followed in order to characterize the performance of T650/MHT-R composites at ambient and elevated temperatures is given.

2.1. Material Systems and Manufacturing

The material systems used in this study were the Thornel® T650/35 8-harness satin woven fabric (370 g/m^3) supplied by Sigmatex (the fibres were produced by CYTEC [9] and featured a 1% UC.309 epoxy compatible sizing) and the thermosetting polyimide NEXIMID® MHT-R which was exclusively been formulated for the HicTac project by Nexam Chemical AB, Sweden (noted MHT-R hereafter) [5,10]. This thermosetting polyimide is made by a combination of 6F-dianhydride (6-FDA) backbone, 4-(Phenylethynyl)Phthalic Anhydride (4-PEPA) end-group crosslinker and ethynyl bis-phthalic anhydride (EBPA) main chain crosslinker [10].

To acquire specimens for the mechanical testing, a 350×350 mm multidirectional laminate with a $[(-45/45)/(90/0)]_{2S}$ quasi-isotropic layup (eight fabric layers in total) was manufactured using Resin Transfel Moulding (RTM) in a stainless steel tool using a flow and pressure controlled injection piston supplied by ISOJET. The curing of the T650/MHT-R laminate was carried out in the following steps: melting and homogenization of the polyimide in the piston at 240°C for 30 minutes; degassing using vacuum at 240°C for 10 minutes; resin injection at 290°C; temperature increase to 340°C at a heating rate of 1.7°C/min, curing at 340°C and 12 Bar for 0.5 h; temperature increase to 370°C at a heating rate of 1.7°C/min, curing at 370°C and 12 Bar for 2 h, cooling down to 80°C during 12 h and demoulding.

2.2. Mechanical Testing

2.2.1. Ambient Temperature

To study the mechanical performance of the T650/MHT-R composites, tensile, compression, and Short-Beam Shear tests were conducted. Tensile testing was carried out on specimens with dimensions 250×25 mm (featuring glass/epoxy bonded with epoxy-based adhesive) according to ASTM D3039 [11]. The specimens were loaded until failure at a constant cross-head speed of 1 mm/min using an Instron 5801 servo-hydraulic machine equipped with a 100 kN load cell while extensometer was used for strain measurement. The stiffness calculation was performed in the elastic region of the stress-strain curve as stipulated by the ASTM D3039 standard (0.1-0.3% strain).

The compressive performance of the T650/MHT-R specimens was assessed using an in-house-built fixture that transfers load through a combination of shear and end loading-i.e. mixed load transfer (MLT) on 80×12 mm specimens (featuring glass/epoxy bonded with epoxy-based adhesive), shown in Figure 1 [12]. The use of this test fixture provides a more convenient way of determining the compressive strength and stiffness in comparison to the classic Wyoming modified ITRII (ASTM D3410) [13], mainly due to the less complex introduction of the relatively small specimen to the fixture, which is an important factor for the high temperature test campaign. Five specimens were loaded at a 1 mm/min rate using an Instron 5801 servo-hydraulic machine equipped with a 100 kN load cell up to catastrophic failure. To measure the strains, 5 mm Kyowa strain gauges (2.09±1.0%

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gauge factor and 120.4±0.4 Ω gauge resistance) were glued on both sides of the specimens. The elastic modulus was determined in the elastic region of the stress-strain curve (0.1-0.3% strain).

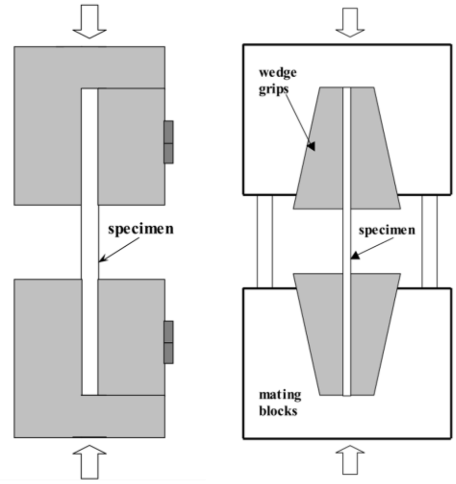

Figure 1. Mixed-load transfer fixture (left) [12]; Wyoming modifiedITRII fixture (right) [13].

For the short-beam shear testing of the T650/MHT-R composites, eight specimens with dimensions 18×6 mm were tested in three-point bending with a 4:1 span-to-depth ratio according to ASTM D2344 using an Instron 5801 servo-hydraulic machine equipped with a 10 kN load cell [14]. The specimens were loaded at a rate of 1 mm/min until a 30% load drop or a two-piece specimen failure occurred or the head travel exceeded the specimen nominal thickness (approximately 3 mm) as stipulated by the standard [14].

2.2.2. Elevated Temperature (320°C)

Considering the inherent difficulties of testing at such high temperatures pertain to specimen gripping and strain measurement, some modifications in the setup for ambient temperature testing (presented in Section 2.2.1.) were done. With regards to specimen mounting and gripping, given that conventional tabs (glass/epoxy bonded with epoxy-based adhesive) could not be used at such temperatures and to avoid drilling a hole to the specimens (potential stress concentration locus). Therefore, it was decided to use dog-bone specimens (lower volume) and to develop a different type of fixture able to perform well at 320°C [11]. In the developed fixture shown in Figure 2, the specimen was clamped using six M6 bolts each tightened to 30 Nm. In that way no specimen slippage ensued.

Figure 2. Developed fixture for tensile testing at 320°C [11].

As for the strain measurement, since the use of strain gauges was not plausible at 320°C due to problems related to the attachment/gluing of the strain gauges on the specimens, Digital Image Correlation (DIC) was utilised (GOM Aramis). The strains acquired by DIC were used for the calculation of stiffness and strain-to-failure of the T650/MHT-R system at 320°C for five specimens.

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It should be noted that a calibration of DIC was carried out prior to testing and a comparison with the strains acquired by the Instron machine and strain gauges suggested an acceptable correlation. The DIC setup used in this test campaign at 320°C is shown in Figure 3.

Figure 3. Setup of the GOM Aramis system for for tensile testing at 320°C [11].

With regards to the compression and Short Beam Shear testing, a similar procedure to that presented in Section 2.2.1 was followed, however in compression the strains were acquired by DIC. Nevertheless, to ensure that the tests were carried out at 320° and no temperature mismatch would occur, thermocouples were used to monitor the temperature at the fixtures while the specimens were kept in the temperature chamber during the entire procedure.

2.3 Microscopy

To investigate the failure morphology of the T650/MHT-R at ambient and elevated temperature optical microscopy was used. Selected specimens from compression and Short-Beam Shear tests, were mounted in Struers EpoFix cold mounting resin and were then ground and polished up to 1 μm grit size using a Struers Labopol-S® polishing machine. Images of failure morphologies were obtained using a Leitz Arisotomet optical microscope, and processed using the Leica Application Suite software.

3. Results

3.1. Mechanical Testing

The mechanical properties of the T650/MHT-R composite system at ambient and elevated temperature are presented and compared in Table 1 while representative stress-strain curves for tension and compression and load-displacement curves for short beam shear are presented in Figure 4, Figure 5 and Figure 6 respectively. Based on the results shown in Table 1 and Figures 4-6, high temperature did not have the same effect on all the mechanical properties of the T650/MHT-R. In particular, in tension the properties (fibre-dominated) were slightly affected, an approximately 9% drop in stiffness and trivial change in strength (within statistical error) was observed. On the contrary, the matrix dominated properties (compression and Short-Beam Shear) were affected to a larger extent; approximately 20% stiffness reduction in compression and 30% strength reduction in both compression and Short-Beam Shear.

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Table 1. Effect of high temperature on the mechanical properties of T650/MHT-R.

Property/Temperature	*RT*	*320°C*
Tension		
Stiffness (GPa)	45.0±0.5	41.2±0.9
Strength (MPa)	488.1±34.6	496.0±17.3
Strain-to-failure (%)	1.3±0.0	1.3±0.0
Compression		
Stiffness (GPa)	46.2±1.6	37.0±0.8
Strength (MPa)	393.0±20.3	272.6±31.6
Strain-to-failure (%)	0.9±0.1	0.8±0.1
Short-Beam Shear		
Strength (MPa)	41.3±1.7	28.1±3.7

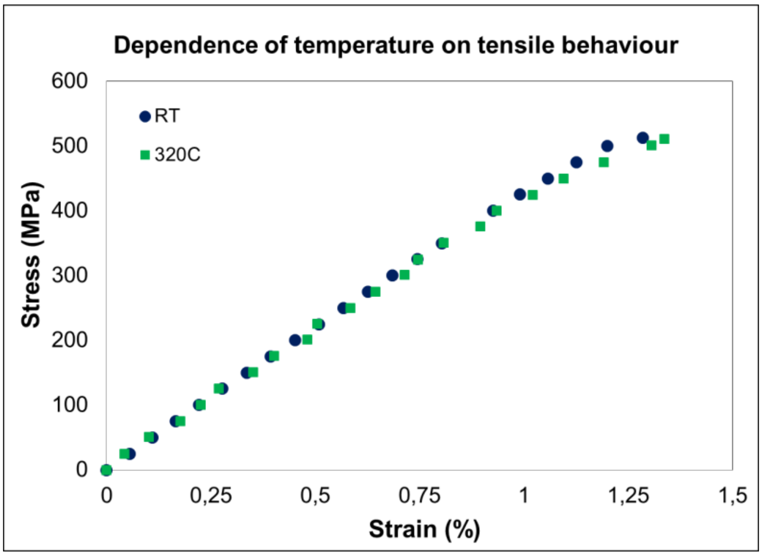

Figure 4. Comparison of representative stress-strain curves in tension.

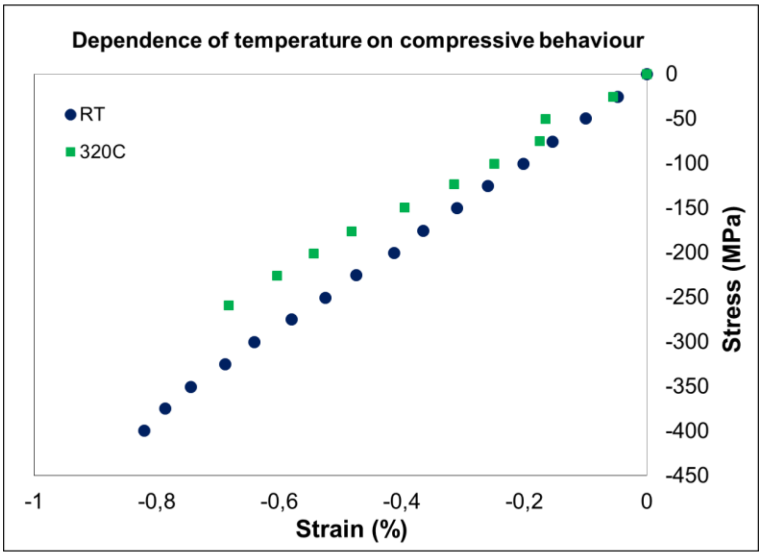

Figure 5. Comparison of representative stress-strain curves in compression.

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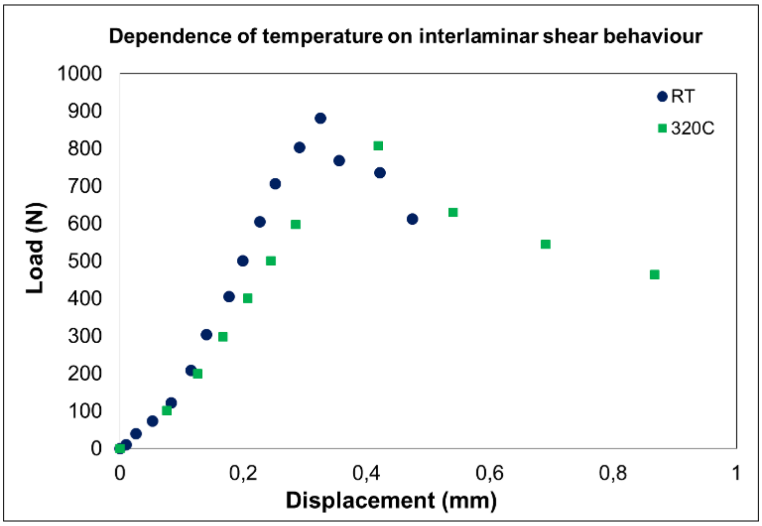

Figure 6. Comparison of representative load-displacement curves in Short-Beam Shear.

3.2 Microscopy

The purpose of the optical microscopy was to provide insight for the decrease by approximately 20% in stiffness and 30% in strength in compression and Short-Beam Shear at 320°C. Compression and Short-Beam Shear specimens tested at ambient temperature and at 320°C were examined using optical microscopy to investigate their fracture morphologies and thus highlight the differences in the failure process and identify the cause of the deterioration in the performance. In Figure 7 and Figure 8, representative fracture morphologies from compression failed samples tested at ambient temperature and at 320°C are presented respectively.

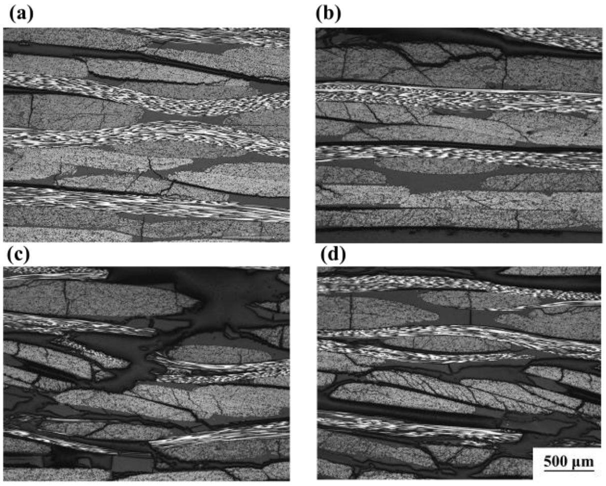

Figure 7. Optical micrographs illustrating representative failure morphologies in compression specimens tested at ambient temperature.

In particular, in Figure 7a and 7b delaminations and matrix cracking/ply splitting as appeared through the thickness as well as their interaction in delamination migration in adjacent ply interfaces are highlighted. In Figure 7c and 7d the fracture morphology at the location where the catastrophic failure occurred is highlighted. Extensive matrix cracking at the off-axis plies and multiple delaminations at various ply interfaces through the thickness are evident especially in the vicinity of the 0° load-bearing fibres, which led to the loss of lateral support and thus to out-of-plane buckling and eventually to

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global failure. On the contrary, the failure morphology at the compression specimens tested at 320°C differed significantly. The failure process was mainly characterized by the delamination shown in Figure 8a which led to the out-of-plane buckling of the 0° load-bearing fibres and as a result to the catastrophic failure (Figure 8b,c,d). These observations indicate that delamination occured rather early in the failure process which impaired the laminate from the load-bearing capability of the 0° fibres and thus caused premature failure. Away from the area where the failure occurred the plies and ply interfaces were intact.

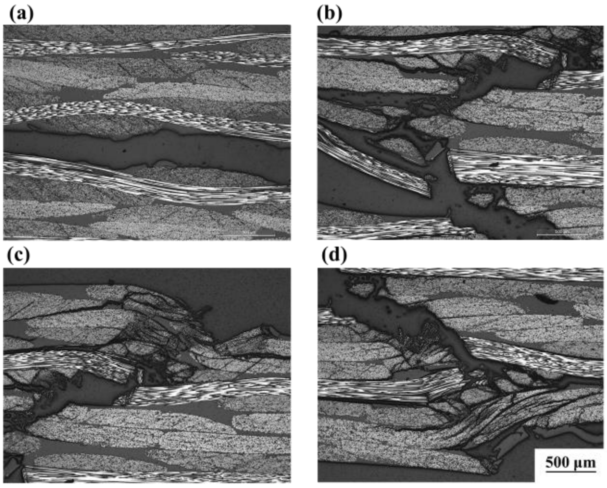

Figure 8. Optical micrographs illustrating representative failure morphologies in compression specimens tested at 320°C.

With regards to the Short-Beam Shear, in Figure 9 representative failure morphologies of specimens tested at ambient temperature (a & b) and at 320° (c & d) are presented. As can be clearly seen the failure morphologies differed. In particular, significantly higher amount of damage can be seen in the specimens tested at 320°C; that is ply splitting at the off-axis plies and extensive delamination especially in the vicinity of the 0° load-bearing plies.

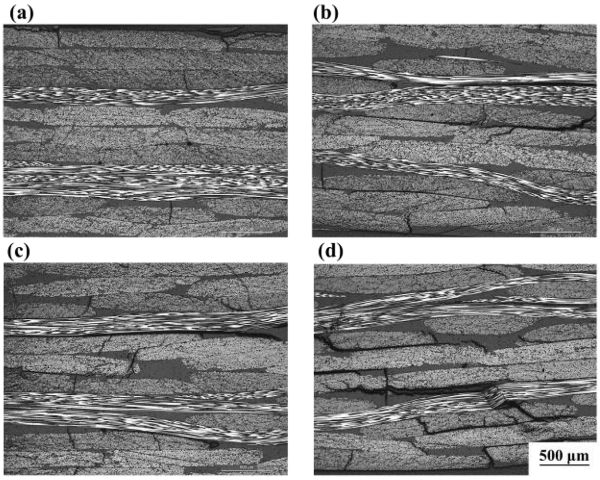

Figure 9. Optical micrographs illustrating representative failure morphologies in SBS specimens tested at ambient temperature (a & b) and at 320°C (c & d).

4. Summary

In this study, the effect of high temperature (320°C) on the mechanical performance of the T650/MHT-R polyimide-based composites was assessed with the aid of tensile, compression and Short-Beam Shear testing. While in tension no effect in terms of stiffness and strength was noted (fibre-dominated property), the high temperature had a more pronounced influence on the compression

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and Short-Beam Shear performance (matrix and fibre/matrix interface-dominated properties), leading effectively to an approximately 20% stiffness reduction in compression and 30% strength reduction in both compression and Short-Beam Shear. To shed light on this phenomenon optical microscopy was employed. The fracture morphologies of compression and Short-Beam Shear specimens tested at ambient temperature and at 320°C were examined and compared in order to highlight the differences and identify the cause of the drop in stiffness and strength. The observations indicated that the exposure to high temperature during mechanical loading affected the stiffness and toughness of the matrix as well as the fibre/matrix interfacial strength which translated to a larger amount of delaminations mainly in the vicinity of the 0° load-bearing plies and matrix cracking/ply splitting at the off-axis plies compared to those noted at the ambient temperature tested specimens. To provide further insight to the behavior of the NEXIMID® MHT-R polyimide resin at high temperatures further delamination fracture toughness tests (DCB) are required. Nevertheless, considering the current outcomes, the T650/MHT-R polyimide-based composites seem to perform reasonably at 320°C.

Acknowledgments

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