Polyimide thin-ply composite

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Abstract:

Mechanical performance of composite structures is influenced by the accumulation of damage from the manufacturing process and throughout the whole service life. For instance, an aircraft is subjected to a combination of mechanical loading and the thermo-oxidative environment from the take-off to the landing. Therefore, this degree project consists of studying the damage initiation and evolution in carbon fibre reinforced polyimide composites and assesses the thickness effect of the laminated composites. After manufacturing, the level of residual thermal stresses occurring at room temperature lead to the occurrence of microcracks in bundles of the quasi-isotropic composites. Further cooling to cryogenic temperature creates new cracks were appearing. This reinforces the conclusion that cracks are created due to thermal stresses. Comparison between a baseline composite made of carbon fibre T650 8-harness satin weave with thermosetting polyimide resin (ply thickness= 190µm) and thin-ply textile laminate made of Textreme carbon fibre IMS65 (ply thickness=83µm) with the same resin shows that the ply thickness has a significant effect on suppressing or delaying the occurrence and the propagation of microcracks after mechanical loading. It is assumed that there are some edge effects leading to different damage state in 90° and ±45° layers.
Acknowledgements:

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I would like to deliver a special thank you to my mom and my girlfriend and Once more - Thank you all for the help.
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1. Introduction

The use of organic polymers-based epoxies or phenolic resin material for the manufacturing of aeronautic and aerospace components is of a great interest thanks to their availability and acceptable cost, whereas for high temperature applications and harsh environments they became unsuitable and degrade rapidly and lose their high performance (a significant drop in of mechanical properties). This lack of performance at high temperature limits the use of those composite materials and their benefits such as high specific stiffness and strength superior than metals and ceramics.

Polyimides have good thermal stability and high heat resistance (Tg above 350°C) which makes them currently the best alternative and the ideal material for severe conditions such as high temperature and pressure applications. When polyimides are combined with carbon fibres, a strong composite material is created that may replace heavier metallic components in such conditions. For instance, aero-engine applications require a composite that will perform in high temperatures because of the air travelling inside the engine and undergoing compression which lead to high temperatures.

Because of this, Polyimides are currently the most commonly resin considered for manufacturing of high performance composite for aeronautical applications with service temperature in the range between 250°C and 300°C [1,2]. High residual thermal stresses develop after the curing process and especially when cooling down to room temperature (RT), it is because of the difference in the coefficient of thermal expansion of the matrix and the fibres. These stresses are of such magnitude that they can cause premature damage formation in the form of multiple cracking within the plies of the composite and hence result in a premature deterioration of the mechanical performance. The initiation and the propagation of intralaminar microcracking has notably attracted researcher attention. Therefore, it was of great interest to investigate the potential of thin-ply composites as a material that may increase the composites general resistance to damage [3] and thereby suppress premature formation of manufacturing induced defects in the form of microcracks due to residual thermal stresses.

Therefore, it was decided to investigate the polyimide thin-ply laminate effect on delaying or suppressing the occurrence and the propagation of transverse cracks due to curing stresses and under quasi-static mechanical loading. This was done by comparing the crack density in a traditional satin weave reinforced polyimide composite with polyimide thin-ply composite. The
comparison was performed by visual inspection of samples using optical microscopy both after the manufacturing process and after quasi-static mechanical loading. Firstly, tracking the evolution of intralaminar cracks after the fabrication process, secondly tracking the formation of cracks under quasi-static mechanical loading as shown in Figure 1.

![Quasi-static mechanical testing flowchart](Image)

**Fig.1. Quasi-static mechanical testing flowchart**

**2. State-of-the-art:**
Composites, an inseparable assembly of non-miscible materials with different structures and properties, are a group of materials where the individual qualities of constituents are combined and complement each other to provide a heterogeneous material with properties that are different compared to the constituents.

Composites have found usage in many structural applications like the automotive field, sporting goods, medicine, construction, ships and before that they dominated the manufacturing of aerospace panels and airframes due to their excellent specific modulus and strength [3].

**2.1. Damage in composites:**
Composite materials and especially carbon fibre reinforced composites (CFRP) used in aerospace grade are exposed to damage by mechanical and thermal loadings throughout their life. Furthermore, researchers classify the damage induced by tensile loadings by three types: transverse microcracking, matrix crack induced delamination and fibre breakage [4]. Generally speaking, transverse matrix cracking takes place as the first damage behaviour through the thickness of the first ply failure, then the delamination damage occurs in the second step and finally the fibre breakage came in the last level of deterioration [3]. What makes things difficult
is the complexity of the damage procedure in composites because it depends on several parameters such as the properties of each constituents, the orientation of fibres, the characteristic and the nature of loading and stacking sequence [3]. For instance, and as a consequence of the quasi-static tensile loading applied in this project, transverse cracks appeared first in the 90° orientation.

2.2. Thin-ply laminate:
There are many studies [3,11-16,18-20] that have studied the influence of ply thickness on damage been done to know how to limit or decrease the damage in composite materials and most of them show that the ply thickness influences each damage mechanism. For instance, in a study carried out by Sihn et al. on thin-ply laminated composites, it was found out that microcracking, delamination and splitting damage are suppressed for thin-ply laminate composites during static, fatigue and impact loading [3]. They performed experiments on laminated composites with the same stacking sequence and volume fraction of each ply orientation, the only difference between them is the number of layers. The results-they have obtained-highlight the effect of number of layers on decreasing the onset of the delamination stress, in other words reducing the ply thickness makes it possible to have a high number of grouped layers which lead to increase the threshold of delamination [7]. Since a laminate-theory based approach does not make difference between laminate with different thickness under the same axial loading, the onset of microcracking damage cannot be shown appropriately by analysing that stress magnitude. Consequently, a probabilistic argument defined bay Rodini and Eisenmann suggest that a thin-ply laminate contains statistically less defects than the thick one [5]. Afterward, Gonzalez et al. established an interesting work and concluded that the thick plies decrease the resistance of damage of each structure [9]. However, in more recent years there has been a study carried out by Camanho et al. analysing the impact of the thickness on the in-situ strengths, likewise on the threshold of the onset of free-edge delamination, it was found out that transverse cracking and delamination resistance are improved with thin-ply laminates [10]. Arteiro et al confirmed that the ply thickness affects the reaction of a material under a mechanical loading [11].

Furthermore, one of the new technologies interested in producing thin-ply laminates with high performance is the tow-spreadning technology which helps on separating the plies with the same orientations instead of grouping the plies [12]; otherwise the thickness of the same kind of the plies will be less [3]. Then laminates with high resistance of microcracking damage and
free-edge delamination will be made, for instance if we consider this following laminate 
$[0_n/\pm 45_n/90_m]_s$ will exhibit a low damage resistance than the quasi-isotropic laminate 
$[(0/\pm 45/90)]_{ns}$ [13]. In addition, thin-ply Non-Crimp Fabrics shows more benefits for laminate 
design, for instance easy laminate homogenization, however in that case stacking plies need 
more time and cost much higher. Moreover, with this tow-spreading technology thin plies 
whose thickness is around 1/3 the conventional ply thickness can be made, the thing that is 
more desirable [14,15].

Early works have been done by S. Sihn et al consisting of many experiments that were set 
up in order to evaluate and to study the effect of reducing thickness on improving the 
performance of tow spread thin-ply laminates. The thickness of the spread thin ply was about 
0.04 mm which is apparently less than 1/3 of the conventional ply of 0.125 mm thick. In order 
to study the thickness effect of the laminated composite, they performed mechanical tests on 
specimens that were made with the same material and the same spread tows but with 
grouped and dispersed laminations of the plies.

Normally, they have thin and thick laminations that were made with the spread thin plies 
technology mentioned before. Moreover, a single ply in the thick lamination is five times thicker 
than a single thin ply [3].

![Fig. 2. Micrographs of test samples made by tow-spread thin plies. a/ thin 
lamination with thinner single ply and b/ thick lamination with a single ply five time 
thicker [13].](image)

In their study, the evaluation of the thickness effect was based on measuring basic elastic 
and strength properties of the quasi-isotropic thin and thick specimens while the presence of 
defect hole or not under static, fatigue and impact loading and comparing them. It is worth 
noting that the stress-strain curves, AE counts, c-scan images and x-ray were used to extract 
experimental observations that confirm the suppression and/or delay of microcracking
damage, delamination and splitting damage within a thin-ply laminates with and without the hole effect.

![Diagram showing the increase of transverse strength of a ply when the ply thickness is reduced.](image)

*Fig. 3. The increase of transverse strength of a ply when the ply thickness is reduced.*

It was noticed from the unnotched test static loading that thin-ply specimen exhibited linear stress-strain relationship during the entire loading, even close to the ultimate failure stress whereas thick-ply specimens exhibited non-linear behaviour. The non-linearity in the latter case was explained by irreversible damage caused by microcracking and delamination. It was also observed that thin specimens exhibited 10% higher ultimate strength values. From stress-controlled fatigue tests it was shown that the thick specimens exhibited an increase in free edge strain due to damage and its influence on stiffness already after 10000 cycles whereas thin-ply ones reveal low or no visible damages even after 50000 cycles of the fatigue loading. It was observed that the residual stiffness and strength of the thick-ply laminates degraded while they degrade negligibly for the thin ones. It was mentioned also that the averages of Young’s moduli dropped by just 2.8% for the thin specimens and 17.7% for the thick ones and their average strength decreased by 16.8% while for the thin laminates it knew an increase by 4.4% which confirm that thin-ply laminates are more durable than the others under the unnotched test loading.

Furthermore, Saito et al. [39] used the energy release rate of the intralaminar transverse cracks and the finite element analysis to study the effect of decreasing the ply thickness on the occurrence and the dissemination of the damage mechanisms with cross-ply laminates under the application of a monotonic tensile loading and it was found out also that when the ply
thickness increases the crack extension is faster, otherwise the propagation of the transverse cracks takes place earlier or in low strains and the strain of its penetration in the 90°layer decrease with the ply thickness. It is worth noting that it was the same depiction for the variation of the normalized crack length in function of the strains.

This ability of thin-ply laminates to suppress or delay the microcracking and delamination damages is also demonstrated by Arteiro et al in an experimental study of plain tensile strength, open-hole and bearing tests [11]. Besides, Boniface set a diagram showing that when the thickness of ply decreases the crack initiation strain tend to grow larger. Otherwise, the transverse cracks will not happen until the strain arrived at 2.00 % of the failure strain with that condition of reducing the thickness sufficiently [16].

2.3. Spread tow technology:

Producing thin-ply laminates needs more precaution to make them without any damages such as filament cut or fluffing because of the strong external forces applied in the conventional processes of ply thinning. More recently Kawabe et al. worked on producing large-tow fibre bundles using a pneumatic tow-spreading technology in order to make thin plies with any deterioration. Consequently, they get extraordinarily thin-layers with a thickness of about 40µm and these thin plies provide excellent mechanical properties of the laminates, for instance the high initiation stress of transverse cracks under tensile loading, the rise in fatigue life and the elimination or delayed of free-edge delamination [24,3]. The industrial Technology Center in Fukui prefecture was the first one who produced thin plies by using tow-spreading technology with the conventional tow for instance 12 K filament tow which is promising and cost-effective way. First of all, the spreading machine has an air duct and a vacuum that absorbs the air downward through the air duct, while the tow passed through the machine and the air flows through the air duct by the action of the vacuum then the tow will sag following the air flow direction and then the tension is lost and provide a tension-free state instantaneous. The spreading of the tow may be kept continuous and stable by using a uniform airflow moderately operating on the tow [3].
Furthermore, damage can be avoided while producing the filament fibres thanks to the velocity of the airflow which is adequately low. Basically, the variation of the airflow’s velocity through both sides near and away of the tow provide a gradient of pressure in that area, besides the pressure close to the tow appear to be greater than away from it. This pressure difference generates an aerodynamic force that leads to the loss of tension by the filament fibres immediately. The Figure 2 demonstrates that the air flows during the tow-spreading and drive up the spreading more and faster.

Fig. 4. schematic of tow-spreading technology by pneumatic method

Fig. 5. schematic of the airflow’s effect in spreading process [13]

It was found out that the tow thickness is small when the tow is wide before spreading and the following Figure 3 shows the difference in tow dimensions before and after the processing [19].
2.4. High temperature composites:

In severe conditions such as high temperatures and high pressure, there is a need for a matrix with high service temperature. There are several matrices that could be used at high temperature such as bismaleimides and cyanate esters but polyimides one of the best choice and ideal for high temperature applications due to their good thermal stability and elevated heat resistance. This makes them the polymer selected for further studies within this investigation. Polyimide is formed after polymerization of imide monomers. The monomers are in themselves a result of reactions between dianhydrides and diamines. The high stability of the systems yields in service temperatures of polyimides between 250 °C and 300°C [26].

For that reason and others, many studies had been carried out to assess the evolution of micro cracks in polyimide composites subjected to mechanical loading and/or thermal cycling [20,21]. In more works, Scola and Wai experienced the effect of ageing at 371°C and different pressures on the degradation of fluorinated polyimides [22]. Besides, the morphology, the stiffness and the strength of the composite is highly affected by thermal cycling and aging at high temperatures [23-25] and other researches investigate the influence of aging and thermal cycling on the mechanical properties and the damage evolution [26-28]. It was found out that the morphology changed and a drop in the mechanical performance was noticed. Moreover, Lafarie-Frenot et al. [26] demonstrated that the propagation of cracks and damage is accelerated by ageing and thermal cycling. In a degree project, McLaren found out that the aging time increased the crack density and thanks to damage and delamination already occurred in aged samples less cracks appear when subjected to mechanical loading. Moreover,
the highest crack density was reached at 90° orientations of the bundles and it accelerate the degradation of the material [29].

G. A. Owens & S. E. Schofield found out that microcracks appeared first in the outer plies due to thermal cycling until achieving a stress-relieved state and then by keeping thermal cycling, cracks appeared in the inner plies [30].

It was shown that microcracking does not affect the fibre-dominated properties whereas the matrix-dominated residual strengths in the inner plies at both room temperature RT(20°C) and 232°C are highly reduced because of cracking. Furthermore, microcracking and reduction of the matrix-dominated properties in the inner plies became more important with the isothermal ageing after thermal cycling [31]. At high temperature, the weight loss and matrix cracking are sharply increased.

More recently, a study carried out by Zrida et al. [31] demonstrates the development of high number of cracks in the quasi-isotropic bundles due to the elevated thermal stresses taken place after a gentle cooling down to room temperature. It was observed that the highest initiated crack density is on the surface +45-layers and that it is larger in 90-layers than in ±45-layers. Besides, the effect of aging on the increased cracks density was shown from the degradation of the elastic and mechanical properties of the composite [32]. Shang et al. confirmed that an increasing crack density lead to decrease linearly the stiffness and the strength of the composite material [33]. Giannadakis et al. have demonstrated that the aging time affect the intralaminar crack density [33].

The difference between the coefficients of thermal expansion of the matrix and the fibre is considered as the primary reason of microcracking of fibre reinforced polymer composites when only curing stresses are present, it is because of the creation of the residual thermal stresses after the cooling down from high processing temperatures.

Regarding the mechanical properties of the T650 (Thornel®T650/35 8-HS woven fabric (370 g/m³))/MHT-R (the polyimide resin NEXIMID MHT-R) quasi-isotropic laminate, they were provided by Tsampas et al [35].

<table>
<thead>
<tr>
<th></th>
<th>E* Tensile [GPa]</th>
<th>E* Compression [GPa]</th>
<th>Tensile strength [MPa]</th>
<th>Compressive strength [MPa]</th>
<th>Strain to failure (in tension) [%]</th>
<th>Strain to failure (in compression) [%]</th>
<th>Fibre failure strain [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>48</td>
<td>45</td>
<td>471</td>
<td>371</td>
<td>1.05</td>
<td>0.86</td>
<td>1.7</td>
</tr>
</tbody>
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*E : stiffness
On the other hand, Varna et al. and Sisodia et al. have described an important phenomenon related to the voids contents, it was found out that the first cracks appear in the layers with voids and by increasing the number of cycles the crack density tend to be similar in all the layers [36,37]. According to Bechel the highest crack density was experienced in surface layers and that on the edge of 90-layer the crack density is higher than in internal 45-layers.

3. Methodology
3.1. Materials:
In this degree project, the material plates were manufactured and provided by Swerea SICOMP using the polyimide resin NEXIMID MHT-R (Nexam Chemical AB) and 8-harness satin weave based on Cytec T650 carbon fibres (Thorne® T650/35 8-HS woven fabric (370 g/m²) was manufactured by CYTEC and provided by Sigmatex) to make the baseline polyimide composite with a symmetrical lay-up [[(-45/+45)/(90/0)]₂s and Textreme carbon fibre fabric IMS65 (IMS65 E23 24K 830 tex by Toho Tenax) to prepare thin-ply textile laminates with the lay-up [[(-45/+45)/(90/0)]₆s. The polyimide composite takes place between the composites designed and intended for severe conditions in aerospace and aeronautics fields thanks to the denoted thermostetting polyimide resin with high thermal stability and ease implementation, it has a low viscosity at the temperature of work because of its low molecular weight and it is 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 cross-linker, this combination enables a high Tg around 372°C [34].

3.2. Manufacturing protocol:
3.2.1. Resin Transfer Moulding:
The use of resin transfer moulding as process of manufacturing is because it is an established method for producing high quality aeronautical grade composites. The method permits high control of high fibre-resin ratio in the completed composite and thereby is a suitable method for production of lightweight parts with high strength. It is commonly used when manufacturing composites for aerospace and high temperature applications. In addition, it can also allow for decorative finish, with controlled fit-up surfaces and it is possible to obtain a surface quality comparable to that of the tool’s surface.

3.2.2. The baseline composite:
The baseline polyimide composite plate used in this study is manufactured and supplied by Swerea SICOMP. It is a quasi-isotropic laminate with a symmetrical lay-up [[(-45/+45)/(90/0)]₂s
and 350 x 350 mm² as dimensions. The manufacturing of the plate has been done using resin transfer moulding RTM in a stainless-steel tool with a flow and pressure-controlled injection piston. First of all, the resin was melted and homogenized at the piston at 240°C for 30 min at 1 bar, then a vacuum was used for degassing at 240°C for 10 min is followed by the injection of the resin at 320°C under 12 bar pressure, the material was cured initially at 320°C for 1h then the temperature went up to 370°C with 1.7°C as a heating rate and the material was post-cured for 2h. Finally, the material was cooled down to 80°C for 12h and demoulded to obtain a laminate with a thickness of 3.04 (3.04±0.03mm), the estimated fibre volume fraction and glass transition temperature are respectively 58% and 370°C [34].

3.2.3. The thin-ply laminate:
Quasi-isotropic thin-ply textile laminate was manufactured at Swerea SICOMP using a high temperature resin transfer moulding, the dimensions of the laminate are 355 x 375 mm² and 24 layers were stacked with a symmetrical lay-up [(−45/+45)/(90/0)]6s. A stainless tool with a flow and pressure-controlled injection piston was used as well. The material was initially cured at 320°C for 2h then post-cured for 1h at 370°C using 15 bar pressures. Finally, the plate cooled down to 80°C for 12h and demoulded then the resulted laminate has 2mm as a thickness and 56% as fibre volume fraction that has been calculated using the laminate thickness and the fabric surface weight.

4. Experimental Procedure:
4.1. Preparation of samples:
Seven (7) samples from thin-ply laminate in Figure 7 and three (3) from the satin-weave laminate were cut carefully using water-jet Discotom cutting machine with a diamond cut-off wheel. In order to prevent the appearance of new cracks after cutting because of the localized mechanical stresses, the cutting machine provide a cooling water system, besides a very slow speed was followed during the cutting process. To recognize easily the samples names and identification numbers were given.
The next step comes afterward is polishing and that is one of the most important preparation steps that would ensure a clear and smooth surface to count cracks and follow the evolution of micro damage. All the specimens were ground using the following sequence of sand papers, P240, P600, P1200, P2500, P4000). Subsequently they were polished up to 0.25 µm grit size with a Struers Labopol-S polishing machine with liquid diamond slurry. The polished edges were inspected regularly with optical microscope to control the polishing quality.

The optical microscope Nikon Eclipse MA200 was behind the investigation and the analyse of damage evolution. A representative length of 50 mm in the centre of each sample edge was inspected, 24 bundles were analysed to determine the initial cracks. The length of 50 mm was changed to 20 mm to save some time and effort during the counting of cracks because of the high number of bundles and to avoid the edge effects. Cracks were counted on both sides specimens for each laminate in ±45° and 90° fibre bundles and the average crack density for each sample was calculated and recorded by dividing the number of cracks counted by the representative length. It is worth noting that some cracks were not taking into account because of their short length (less than one-third the height of the bundle). The following figure shows a standard crack can be counted.
Fig. 8. Micrograph at the edge of one sample from the conventional laminate taken at 10x magnification showing a crack in the 90° fibre bundle highlighted

Tabs were prepared using glass fibre/epoxy, the Araldite 2 was used as an epoxy adhesive to attach them to the specimens. Finally, small sandpaper pieces were attached to the specimens to provide a high friction surface for the extensometer.

4.2. Experimental Conditions:
4.2.1. Quasi-static tensile tests:
Samples were re-polished before starting the tensile tests to minimize any influence of edge effect cracks possibly created as a consequence of additional residual stresses associated with the release of constraints during the cutting and making specimens from the initial plate.

In order to calculate the E-modulus three samples from the thin-ply laminate were tested using the Instron 3366 tensile machine with a load cell of 10kN at LTU. The test method has one step of loading until 0.35% as a maximum strain level followed by one step of unloading until zero load. Measuring samples dimensions (see Appendix) was the first step before mounting them. Then with the cross section calculated and the data collected after each test, additional quasi-static tensile tests at different strains level were performed at Swerea SICOMP (Piteå, Sweden). This time, an Instron 3366 with a load cell of 100kN was used to carry out stepwise loading tests. An extensometer with 50 mm length was mounted and a relevant test method was run on InstronBlueHill software connected to the machine. Besides, the test method has 4 steps, it started by a position step and a loading to the desirable maximum strain level followed by an unloading step which is basically unloading the sample back to zero load.
where the test was started from and finally a position step. When the test is done, the sample was removed from the tensile machine grips and taken to the optical microscope for inspection and quantification of micro damage. Then the same sample, once it has been inspected from both polished edges, is again mounted in the test machine and loaded to the next higher strain level and then unloaded and taken for inspection and damage quantification under an optical microscope. It is worth noting that the samples were loaded to the following maximum strain levels: 0.6%, 0.8%, 1.00% and 1.2%.

5. Results:

The following section of the current report has been divided into two sub categories, analysing and comparing two different polyimide composites (thin-ply and conventional laminate) after the manufacturing processing and mechanical testing. Inspecting samples and counting cracks in section 5.1 have been done to assess the curing stresses effect on damage and how reducing the ply thickness influence laminate resistance towards transverse cracking. Section 5.2 shows the difference in stiffness and its progression after mechanical testing. The occurrence and the propagation of microcracks after mechanical testing is affected by the thin-ply resistance.

The polyimide composite has been tested under quasi-static mechanical loading to induce damage within bundles and assess microcracking damage resistance thanks to the thin-ply laminate and how the performance is improving.

5.1. Curing thermal damage:

Six (6) samples were analysed, three (3) samples from the thin-ply laminate and three (3) samples from the baseline laminate. The cracks generated after manufacturing were reported for each laminate and the following graph in Figure 9, was drawn to compare microcracking resistance in both materials.
Fig. 9. Average crack density of each sample from both laminate after manufacturing

From Figure 9 it is seen that after manufacturing, the thin-ply laminate showed a low crack density comparing to the baseline laminate. The crack density does not exceed 0.15 crs/mm in the thin-ply laminate after the fabrication process.

Fig. 10. Micrograph taken at 10x for one sample from the conventional laminate after manufacturing
**Fig. 11.** Micrograph taken at 30x for one sample from the conventional laminate showing crack and delaminations

**Fig. 12.** Micrograph taken at 10x for one sample from the thin-ply laminate after manufacturing
Micrographs in Figures 10-13 taken at different magnifications for samples from both laminates after manufacturing process showed the presence of voids with different sizes. Cracks and delaminations were clearly shown in the baseline laminate whereas few cracks were found within the thin-ply laminate.

5.2. Mechanical testing:

5.2.1. Stiffness:

Quasi-static mechanical tests were performed to measure and calculate the stiffness in the thin-ply laminate presented in Table 2 whereas the stiffness of the baseline polyimide laminate (E=45GPa) was imported from the literature [34]. Tensile modulus and volume fraction for carbon fibres used in both laminates was reported in Table 3 to compare the difference in the stiffness. Besides, the stiffness progression of the thin-ply laminate after the mechanical testing was presented in Figure 16.
Table 2. The measured and the average stiffness of 3 samples from the thin-ply laminate after loading to 0.35% strains

<table>
<thead>
<tr>
<th>Sample</th>
<th>Tensile stiffness (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>49.6</td>
</tr>
<tr>
<td>S2</td>
<td>50.6</td>
</tr>
<tr>
<td>S3</td>
<td>49.4</td>
</tr>
<tr>
<td>Average</td>
<td>49.9</td>
</tr>
</tbody>
</table>

Table 3. Tensile modulus and volume fraction for carbon fibres used in the conventional laminate and in the thin-ply laminate

<table>
<thead>
<tr>
<th>Fibres</th>
<th>Composite</th>
<th>Baseline laminate</th>
<th>Thin-ply laminate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon fibres</td>
<td>T650/35</td>
<td>IMS65</td>
<td></td>
</tr>
<tr>
<td>Fibre volume fraction (%)</td>
<td>58±1</td>
<td>56±1</td>
<td></td>
</tr>
<tr>
<td>Fibre tensile modulus (GPa)</td>
<td>255</td>
<td>290</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 16. Stiffness progression with strain for the thin-ply laminate samples

5.2.2. Mechanical damage:

Mechanical tests have been performed on samples from the thin-ply laminate to determine the stiffness and its progression in Figure 16. At the same time, the crack density was determined and reported in Figure 17, whereas results for the baseline laminate were imported from literature in Figure 18, to compare effect of the ply thickness on microcracking resistance.
Fig. 17. Average crack density in samples from the thin-ply laminates after loading to specified strain.

Fig. 18. Average crack density in different bundle orientations within one layer from a conventional polyimide laminate after loading to specified strain [38].

From Figures 19-21, micrographs of some samples from the thin-ply laminate were taken at different magnifications after loading to specified strain. Few microcracks took place and no delaminations were observed.
Fig. 19. Micrograph taken at 20x for a sample from the thin-ply laminate after loading to 0.8% strains.

Fig. 20. Micrograph taken at 30x for a sample from the thin-ply laminate after loading to 0.6% strains.
6. Discussion:

6.1. Curing thermal damage:

As mentioned before the inspection of mesostructured and the initial damage state of all specimens from both laminates have been done using the optical microscope before any mechanical testing.

The first cracks were generated because of thermal shrinkage of the resin and residual stress from curing dominating the cracks initiation in the bundles when the composite was cooling down after manufacturing. It is assumed that the difference in the thermal expansion coefficients of fibre and matrix and the weave of the fibre fabric are behind these stresses. As shown in Figure 4, the highest initial crack density was found in the baseline polyimide laminate and it was clear that 0-340=340°C is the difference temperature (340°C is the maximum curing temperature and 0°C the minimum temperature reached in cooling down) causing the damage occurrence after manufacturing.

A very low number of cracks were found in the thin-ply laminate samples and it was observed that most of these cracks are in the 90° layers. The finite element analysis performed by Zrida et al. [31] it was found out that the transverse stresses are amplified by approximately 20% in 90°layers. This is because of a new stress state that is generated once the boundary is
“released” by the cutting process. It is worth noting that the cutting of samples was done at room temperature which was about 20°C which is higher than the lowest temperature during manufacturing, in other words the thermal stresses in the plates were lower by 6-7% but the transverse stresses generated by making new specimens were much larger. Therefore, it was assumed that new cracks appearing in the edge regions of 90° layers are because of the new stress state and that most probably these cracks are not growing inside the specimen where the stress conditions were not affected by the cutting process. Besides, the localized mechanical stresses applied by the cutting wheel can also create defects that serve as crack initiation locations. To confirm or deny this hypothesis of the new stress state, two samples from the thin-ply laminate were re-polished and 9 mm of the material was removed then the representative length was inspected using the optical microscope and no microcracks were found in all the 90° layers which confirm the latest hypothesis.

The ply thickness seems having a high effect on the occurrence and the propagation of damage within the bundles because of the suppression of microcracks in the thin-ply laminate after manufacturing.

In other words, the thin-ply textile showed a considerable resistance to the residual thermal stresses produced by manufacturing processing whereas the crack density in the conventional polyimide composite was much greater which means its low damage resistance because of residual thermal stresses.

Voids were shown in the conventional laminate in Figure 10. They provide dual roles, as was mentioned by Varna et al. and Sisodia et al [36], it was clear to see that some cracks appear at the voids as stress concentrators. Additionally, it was observed that voids prevent the propagation of microcracks and delay the appearance of new cracks by providing a larger stress relaxation.

Delaminations were also observed after fabrication processing of the baseline polyimide composite in Figure 11 and some cracks are likely joining up those delaminations instead of propagating through bundles since a low resistance was depicted and cracks follow the weakest path.

6.2. Mechanical testing:
   6.2.1. Stiffness:
   The Young’s modulus for the conventional laminate T650/MHT-R that was established in the previous work by Tsampas et al. [34] was approximately 45 GPa and the modulus value
determined and recorded for the polyimide thin-ply laminate IMS65/MHT-R was around 50 GPa. The difference in the Young's modulus is explained by the difference in type of carbon fibres and volume fraction; the tensile modulus of T650 carbon fibres is about 255 GPa with 58±1% as fibre volume fraction whereas it is approximately 290 GPa for IMS65 carbon fibres used in thin-ply laminate with 56±1% as fibre volume fraction were reported in Table 3.

As expected it was found out that the stiffness of the specimens was decreasing after each loading cycle during tensile tests because of the evolution of the crack density in the bundles.

6.2.2. Mechanical damage:

Before quasi-static mechanical testing, the samples were re-ground and re-polished and around 9mm of the material was removed from the sample edges to remove the additional cracks appearing in 90° bundles because of the cutting processing, after polishing all the cracks disappear.

Mechanical damage assessment showed that crack density and degraded areas were all present in the baseline laminate samples which explain the lower value of around 45GPa. For some samples, contrary to expectations the E-modulus was showing a higher value after mechanical testing and it was suspected to be an error in operating (mounting samples for instance). Therefore, re-testing samples have been done. Therefore, the stiffness of the thin-ply laminate showed a slight decrease with the increasing loading and the evolution of damage.

It was found out that the first few microcracks appeared within 90° fibre bundles in the thin-ply laminate samples when the maximum strain 0.6% was reached, the increase of loading was resulting in an increase in crack density. The highest crack density was around 3.5 cr/mm, it was achieved at 1.2% maximum strain whilst a crack density of around 2.5 cr/mm was found at 0.42% maximum strain in the conventional laminate. Besides, some cracks start appearing in ±45 thin-ply laminate fibre bundles at high strain level and few cracks propagate throughout two bundles. Moreover, a similar crack density was found for both surface and inner layers in thin-ply laminate samples because it is assumed that there are (generally) no areas that are significantly weaker than others. Besides, microcracks followed a path amongst the fibres and where the mechanical stresses were higher the cracks propagate horizontally throughout the poorest interfacial bonding.

One can notice from the graph in Figure 18 showing the crack density after mechanical testing in the conventional polyimide composite reported by McLaren [38], that a high crack density was found at around 0.42% maximum strain, whereas no microcracking happened.
within the thin-ply laminate at that strain. Furthermore, a low crack density was found at 0.6% maximum strain which explains and confirm the ability of the thin-ply laminated composite in suppressing cracks and delay the propagation of cracks after quasi-static mechanical tests. It is worth noting that some failures were observed in 0° fibre bundles in the thin-ply laminate after the maximum strain 1.2% but no delaminations or laminate failure took place.
7. Conclusion

In this study, it is possible to conclude that the tested polyimide thin-ply laminate showed a high resistance to microcrack formation due to residual thermal stresses generated after the high temperature manufacturing processing. This work confirms the high effect of reducing the ply thickness in increasing the microcracking formation resistance; likewise, the thin-ply laminated composites can suppress microcracking and delaminations due to curing stresses or at least delaying the propagation of microcracks.

It was observed that mechanical loading increases the number of cracks, but polyimide thin-ply laminate showed a high resistance to microcrack initiation and propagation under quasi-static mechanical loading when a low crack density was found at 1.2% as a maximum strain whilst a higher crack density in the conventional polyimide composite was found even at low strain. The orientation containing the maximum crack density from mechanical loading is 90°.

The thin-ply laminate showed a slightly higher stiffness, despite lower $V_f$, because of the higher modulus carbon fibres used in the thin-ply reinforcement. Thin-ply composite combines a high resistance towards curing and mechanical stress-induced microcracking with a high tensile modulus and it can be said that this composite has potential for many applications.
8. Future work
More work is required to reach a well understanding of the material behaviour under thermal and mechanical loading and establish optimum parameters processing to produce the polyimide composite with high quality. Furthermore, this degree project is a preliminary part of a fuller investigation into this polyimide composite with thin-ply laminate and its variables and as a further investigation, the study of their performance in mechanical fatigue, impact loadings and short-beam shear (SBS) tests. Besides, it will be interesting to assess to evolution of damage and the degradation of the laminate after ageing and thermal loading of the polyimide thin-ply composite.
9. References:

19. assan M. EL-Dessoukya, b,fl, Carl A. Lawrence Advanced Composites Research Group, Centre for Technical Textiles, School of Design, University of Leeds, Leeds LS2 9JT


Bowles DE, Shen J. Thermal cycling effects on the dimensional stability of P75 and P75 T300(Fabric) hybrid graphite/epoxy laminates. In: 33rd international SAMPE symposium; March 7–10, 1988.


Appendices:
Appendix A: Equipment List

Samples cutting
✓ Discotom 100

polishing
✓ Buehler MetaServe 250 Grinder-Polisher
✓ Struers LaboPol-5
✓ Kemet liquid diamond

Microscopy
✓ Nikon Eclipse MA200

Tensile Testing
✓ Instron 3366, 10-100kN load cell
Appendix B: Tables of Crack Densities

Table 4. curing damage: Average crack density for each bundle orientation

<table>
<thead>
<tr>
<th>Orientation (°)</th>
<th>Thin-ply laminate</th>
<th>Conventional laminate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>S1</td>
<td>S2</td>
</tr>
<tr>
<td>± 45</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>90</td>
<td>0.14</td>
<td>0.09</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 5. mechanical damage: Average crack density for each bundle orientation

<table>
<thead>
<tr>
<th>Orientation (°)</th>
<th>Strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.35</td>
</tr>
<tr>
<td>± 45</td>
<td>0</td>
</tr>
<tr>
<td>90</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>