AN INVESTIGATION OF THERMOPLASTIC TAPE-TO-TAPE BONDING USING XENON FLASH LAMP HEATING

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Keywords: Thermoplastics, Xenon flash lamp, Tape bonding, Peel strength, Welding

ABSTRACT

This research was undertaken to investigate factors affecting the tape-to-tape bond strength of thermoplastic composite tapes in the Automated Fibre Placement (AFP) process using a xenon flash lamp as the heat source. Two variants of flash lamp were studied with different spectral outputs.

The effect of exposure to the broadband output of the flash lamp on the polymer chemistry of representative thermoplastic tapes was investigated by analysing exposed tape samples. Particular focus was given to the potential for the UV wavelengths within the spectrum to photodegrade the long polymer chains into lower molecular weights, reducing their ability to form strong tape-to-tape bonds. Three spectroscopy techniques were used: Fourier-Transform Infrared Spectroscopy (FTIR); Raman Spectroscopy; and X-Ray Photoelectron Spectroscopy (XPS). The study included carbon fibre tapes with polyaryl ether ketone (CF-PAEK) and polyamide 6 (CF-PA6) matrices. From these spectroscopy studies, no clear evidence was found that exposure to flash lamp energy causes photodegradation of the polymers.

The study then investigated how the spectral output of the flash lamp affected the peel strength of the tapes through peel testing of bonded tape pairs and imaging of the peeled surfaces. Tapes were bonded (welded) by a novel process, using the flash lamp's ability to heat and apply compaction force onto the tapes simultaneously. The peeled surfaces revealed the failure mechanism of the polymer at the interface via Scanning Electron Microscopy (SEM), which in all cases showed ductile failure of discrete polymer regions and not overall brittle failure. The difference between low and high peel strengths was observed to be related to the number of discrete areas of polymer that undergo plastic deformation during the peel.

1 INTRODUCTION

Research into efficient manufacturing processes using high-temperature thermoplastic composites has been ongoing for many years ([1], [2]), but their adoption for applications in the aerospace and automotive industries has been relatively slow, in spite of their clear advantages over thermoset composites through enhanced material properties, faster manufacture and their potential to be recycled. One of the reasons for this delay in adoption has been the challenges inherent in fast, automated tape lay-up processes such as AFP or Filament Winding (FW) to achieve full, in-situ consolidation in a single manufacturing step. These fast processes typically produce partial bonding of the composite tapes, which then necessitates a secondary (slow) heating step in order to reach the desired lay-up quality. These processes also typically induce very fast cooling rates in the processed materials, which limits the level of crystallinity that can be recovered in semi-crystalline thermoplastic polymers, resulting in lower mechanical properties of the part ([2]).

There is a compelling need to understand the tape-to-tape bonding process more closely, especially when using high-power heat sources such as lasers and xenon flash lamps. With a deeper understanding of the behaviour of the thermoplastic polymers during the heating and cooling phases, it may be possible to adapt current automated manufacturing processes to achieve higher tape-to-tape bond strengths and higher quality parts. This paper investigates aspects of xenon flash lamp heating that affect the bond strength of thermoplastic tapes during AFP.

Xenon Flash Lamp Heating

The xenon flash lamp heating system consists of a xenon-filled flash lamp (a quartz glass tube with metallic electrodes), within which an energetic plasma is created in short duration pulses that emit photons from the lower ultraviolet (UV) wavelengths to the higher infrared (IR) wavelengths. The lamp is housed within a larger diameter quartz glass flow tube to allow an annular flow of deionised water to pass the lamp for cooling purposes.

The material of the lamp (or flow tube) can either be clear fused quartz (CFQ), which causes no attenuation across the spectrum, or cerium doped quartz (CDQ), which attenuates the majority of the energy within the UV spectral region. Typical spectral outputs of the CFQ and CDQ variants are shown in **Figure 1**, differing only in wavelengths below 400nm.

The flash lamp and flow tube are contained within a small, T-shaped head with a shaped reflector behind and an aperture in front of the lamp to guide the energy into a solid quartz light guide (**Figure 2**). The light guide acts to guide the diffuse flash lamp energy to the target by means of total internal reflection. The output face of the light guide can be kept as a flat surface or shaped with planar facets to create a desired heating profile ([3], [4]). The output from the light guide is thus high-energy, pulsed, broadband and diffuse.

Three pulse parameters: pulse voltage (typically 150-250V); pulse frequency (number of pulses per second – typically 60-100 Hz); and pulse duration (typically 2-5ms) are varied to modulate the average output power of the system. The maximum average power of the system used in this study was 6kW, with an instantaneous, in-pulse power of up to 85 kW. In the typical AFP usage, where the flash lamp moves on a robotic arm, pulses superimpose as the lamp passes across a moving target, resulting in an approximately constant surface temperature.

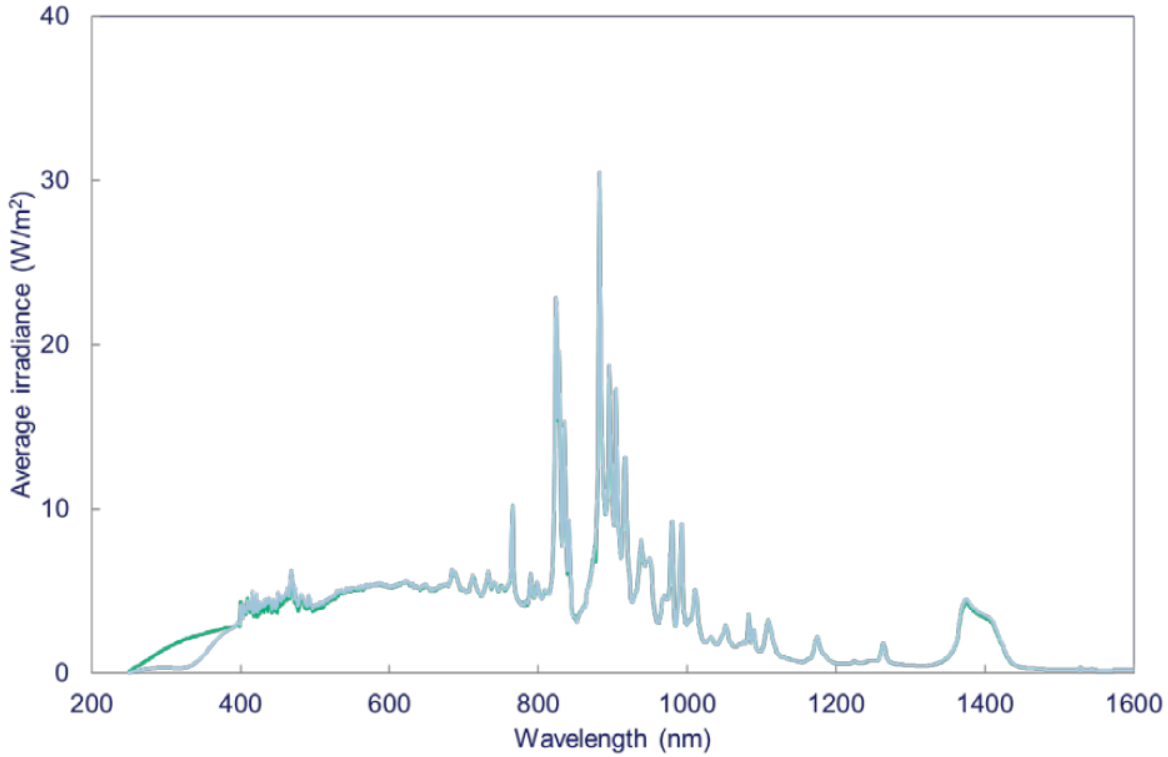

Figure 1: Spectral output of flashlamp with CFQ and CDQ flow tubes. The spectral outputs are identical above 400nm, but below 400nm most of the UV energy is removed by the CDQ case.

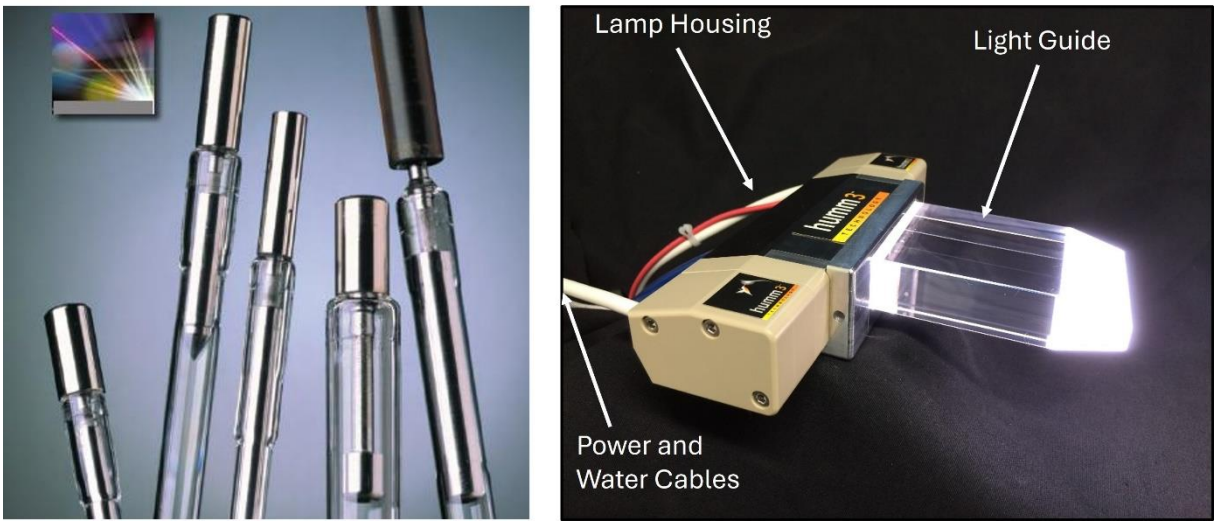

Figure 2: Image showing typical xenon flash lamps (left) and the flash lamp system head (right)

The motivation for this project arose from the results of previous unpublished work carried out at the National Composites Centre (NCC), UK, on AFP of CF-PAEK tapes using the flash lamp. The CFQ and CDQ variants of the system were compared as heat sources on a Coriolis AFP robotic system under nominally identical conditions. It was found that the CDQ variant produced tape pairs with a significantly higher bond strength than the CFQ variant, even though speed, compaction pressure and nip point temperature were matched. It was hypothesised that the additional UV content in the CFQ spectral output may have caused the lower tape-to-tape bond strengths. This motivated the current study to look for changes to the polymer surface chemistry caused by flash lamp irradiation.

It is generally well understood that polymers are sensitive to UV exposure, and can suffer degradation effects such as photooxidative aging, which may result in the breakage (scission) of polymer chains and a reduction in average molecular weight ([5]). However, the main body of literature has looked at long-term UV aging, for example by sun exposure, rather than the short exposure times (fractions of a second) typical of AFP processing.

As a result, this project has tested the hypothesis that changes to the polymer chemistry of the tape surfaces due to exposure to the broadband flash lamp radiation cause a reduction in bond strength. As a first step in this investigation, several spectroscopy techniques were deployed to look for changes to the polymer chemistry due to flash lamp exposure.

2 FLASH LAMP TAPE EXPOSURE METHOD

In the AFP process, each discrete area of the incoming tape is exposed to flash lamp energy for a short time as it passes through the heated zone, before reaching the AFP nip point and being compacted onto the substrate by the roller ([3], [4]). Under typical AFP conditions, the tapes are estimated to be within the heated zone for 0.5s to 1s, depending on the lay-up speed and dimensions of the heated zone. In order to replicate this tape exposure and investigate any changes to the tapes caused by the flash lamp heating, samples of the tape materials were exposed to flash lamp radiation on a moving linear stage without further processing. The exposure rig is shown in **Figure 3** showing the flash lamp light guide directed vertically downwards, heating the tape as it passes on the linear stage. The offset distance between light guide and tape was 20mm.

Tape surface temperatures were maintained at approximately the melting points of the polymers (220°C for PA6 and 300°C for PAEK) by modulating the tape speed at constant flash lamp pulse parameters of 175 Volts, 60 Hertz and 2 milliseconds. A thermal camera was used to monitor tape temperatures during the exposure. Tape speeds of 50mm/s were used for CF-PA6 samples and 40mm/s for CF-PAEK samples. The CFQ variant of the flash lamp system was used to create exposed tape samples for spectroscopic testing. Exposed samples were compared with unexposed (control) samples. All tapes were 1/4” wide.

Additional exposed samples were created using the same configuration but with the CDQ flash lamp variant. These samples were used in the subsequent testing of the strength of bonded tape pairs, described in section 4.

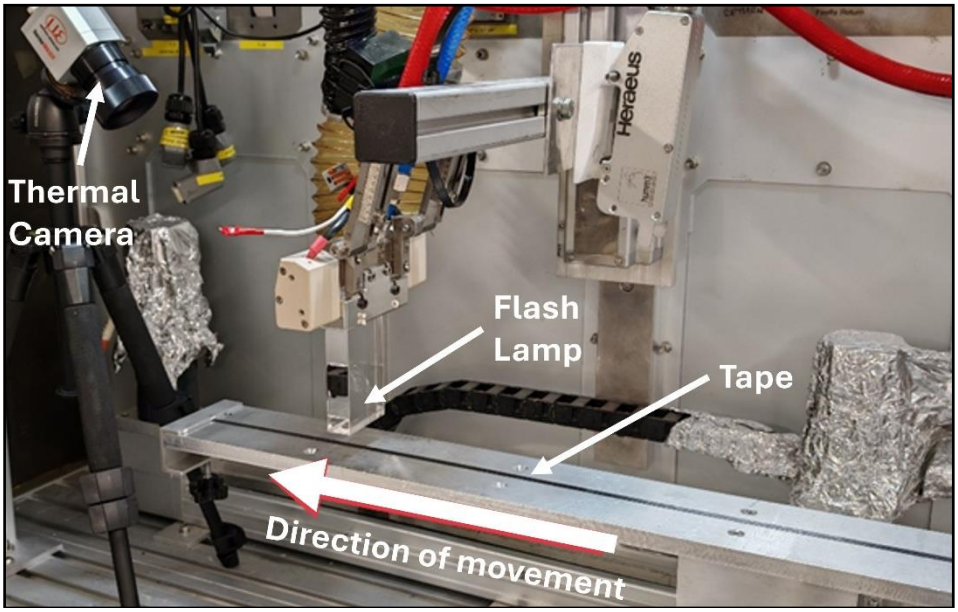

Figure 3: Flash lamp and linear stage used to expose tape samples to representative flash lamp heating

3 SPECTROSCOPY RESULTS

FTIR Spectroscopy Results

Fourier-transform infrared spectroscopy (FTIR) is a chemical analysis technique that applies infrared light to a sample to gain insights into its chemical structure. The FTIR analyses described here were conducted at the NCC. In these analyses, an attenuated total reflectance (ATR) sampling head was used attached to an Agilent Exoscan 4100 FTIR. Spectra were taken between wavenumbers of 600 cm^{-1} and 4000 cm^{-1}. For each material, the control (unexposed) and CFQ-exposed samples were compared. The tapes were held onto the ATR sampling head with a clamp and spectra were taken at three points along the surface of each tape.

Polymer chemistry changes to the tape surfaces were expected to be evidenced by changes to spectral peak positions and heights, or the appearance of additional peaks. Representative results are shown in **Figures 4** and **5**. A typical FTIR spectrum is shown in **Figure 4** for a control sample of the CF-PA6 material**.** The peaks have been annotated with their corresponding chemical bonds within the thermoplastic resin.

Figure 5 shows a comparison of nine FTIR spectra from control and CFQ samples of CF-PAEK tapes. No clear changes to peak positions, heights or numbers are observed.

It is worth noting here that carbon fibre is highly infrared absorbent across the full wavenumber range. Slight changes in the resin-to-fibre ratio on the tape surface will alter the baseline height of each graph. This effect is known as baseline drift. The slight peak height differences seen in **Figure 5** are attributed to this baseline drift rather than any chemical changes.

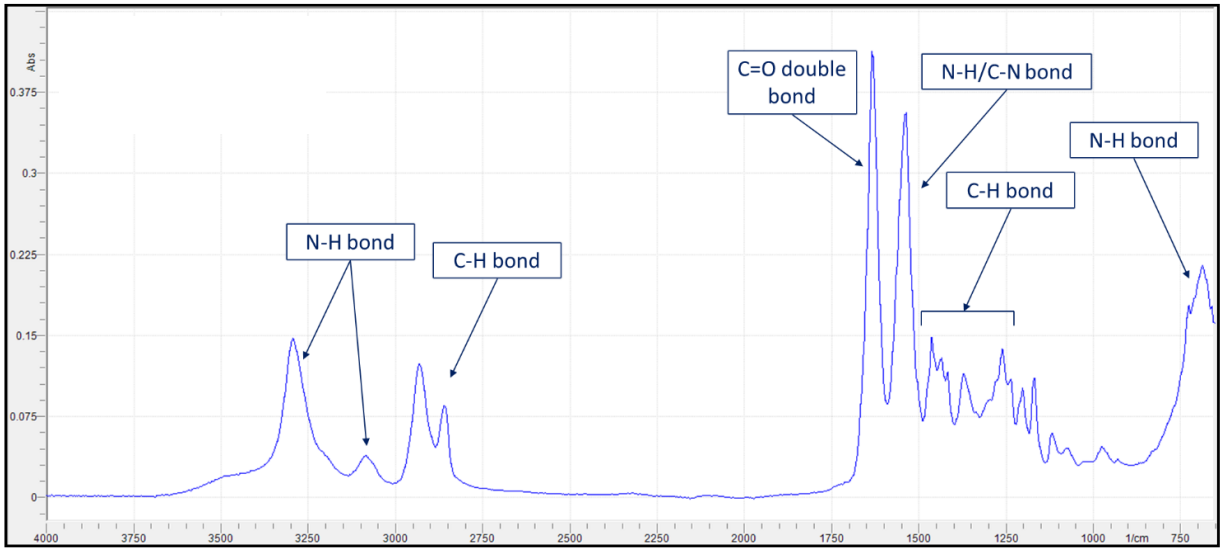

Figure 4: FTIR spectrum of unprocessed CF-PA6 tape, 650-4000cm^{-1}.

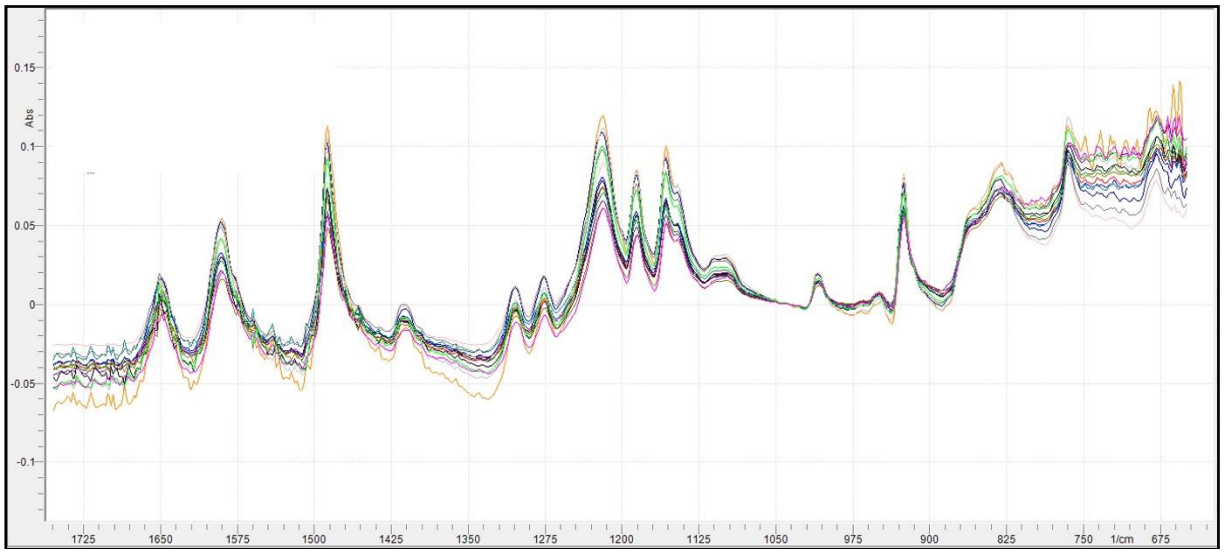

Figure 5: FTIR spectrum of control and CFQ LM-PAEK tapes, 650-1750cm^{-1}

The results shown in Figure **5** are typical of all other FTIR analyses conducted in this project. No changes to spectral peaks were observed in the case of the CF-PA6 samples. It can be concluded that no evidence of chemical degradation was observed by FTIR spectroscopy when the CF-PAEK and CF-PA6 materials were exposed to flash lamp radiation.

Raman Spectroscopy Results

Raman spectroscopy was chosen as an alternative chemical analysis technique for composite materials that typically applies high-intensity laser light to a sample to gain insights into the chemical structure. It was deployed in this project as a complementary technique to FTIR to look for evidence of chemical degradation. The Raman analysis was conducted at the University of Bristol. For all the samples tested, significant baseline drift was observed, which affected the ability to interpret results. One possible explanation of this baseline drift is the autofluorescence of exposed carbon fibres at the surface. The Raman and FTIR spectra of a control CF-PA6 tape sample are overlayed in **Figure 6**. The Raman trace is in black and the equivalent FTIR trace is in red. The peaks in the Raman trace are poorly defined and the baseline drift masks the lower intensity peaks between 1000-1750 cm^{-1}. The fluorescence

was even more pronounced for the CF-PAEK material samples tested, so that only the most intense peaks could be seen. This meant that no comparison between samples could be made as it was not possible to assess whether peak heights were changing or if new peaks were appearing.

Within the budgetary and time constraints of this project, it was not possible to improve this method. For this reason, Raman spectroscopy was discounted from the rest of the study.

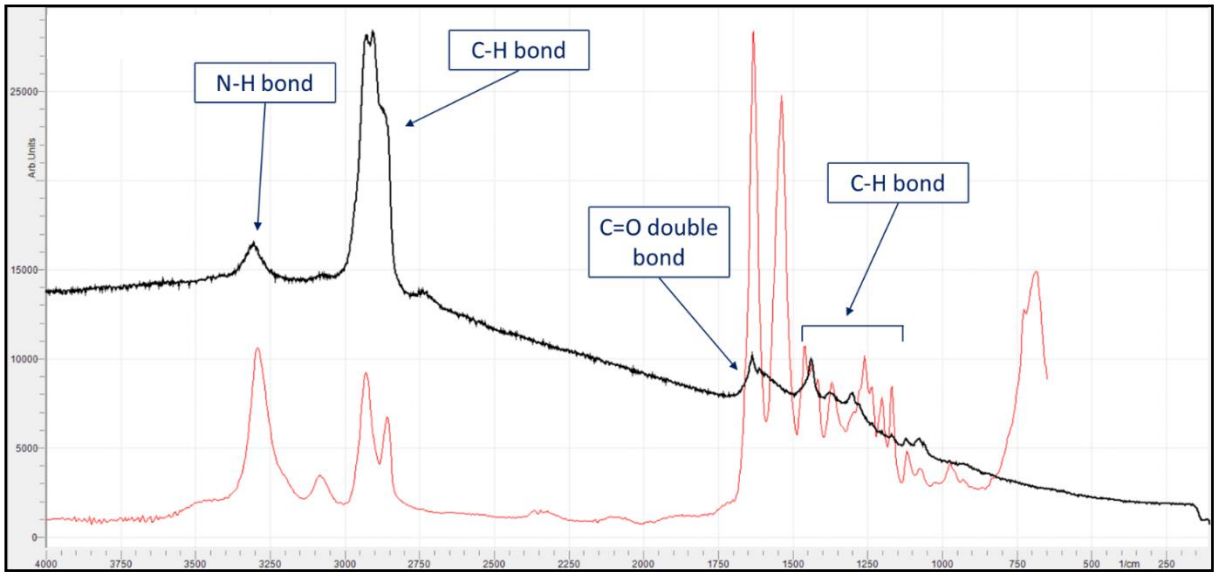

Figure 6: Black trace – Raman spectrum of control CF-PA6 tape. Red trace - FTIR spectrum of control CF-PA6 tape.

X-Ray Photoelectron Spectroscopy Tests

The results from FTIR and Raman spectroscopy did not show any clear evidence for polymer degradation of the composite sample surfaces. However, both techniques assess a relatively thick (several micron) surface layer of the material. It was thought that the high energy pulses from the xenon flash lamp may not penetrate into the material to this depth, and thus these two spectroscopy techniques may not be able to distinguish surface effects from the unaffected bulk material. As an alternative technique, X-ray photoelectron spectroscopy (XPS) was explored.

XPS is a spectroscopic technique that applies x-rays to the surface of a sample and measures the kinetic energy of the electrons that are emitted from the surface. It provides information about the chemical elements present in the sample and is a very surface-sensitive techniques that is only influenced by the top 10 nm of the material ([6]).

The XPS tests were carried out by Harwell XPS, at the EPSRC National Facility for XPS in Oxfordshire, UK. Technical details of the XPS set up and analysis are not provided here (but can be supplied by contacting the author). The samples tested were the control, CDQ and CFQ samples of each of the CF-PA6 and CF-PAEK tapes. Five repeats were carried out for each sample type, and particular interest was paid to any change in the oxygen percentage calculated, as this could be an indication of oxidative degradation of the polymer.

Composite materials are, by their nature, more difficult to assess than single material systems, especially as the ratio of resin and fibre on the surface of the material may vary with surface location and change during processing. If the ratio of resin to fibre changes, then the elemental ratios will vary. This appears to be the case for the CF-PA6 samples analysed here. As shown in

Figure **7** (left graph), the carbon percentage for the CF-PA6 tapes increased from 79.5% in the control samples to 81% for the CDQ samples and 83% for the CFQ samples. This may suggest an increase in fibre on the surface of the CDQ and CFQ samples as the carbon fibre contains solely carbon atoms compared to the polyamide resin which contains carbon, nitrogen and oxygen. A corresponding drop in nitrogen and oxygen percentage (Figure **7**, middle and right graphs) was observed. If oxidative degradation of the polymer had occurred, then an increase in the percentage of oxygen on the tape

surface would be expected. However, any increase in oxygen percentage from oxidative degradation may be masked by changes to the resin to fibre ratio. Hence it was not possible to make a firm conclusion that there was evidence of oxidative degradation. It should be noted that the standard deviation on all results (shown as black vertical bars in the figure) was large and therefore further testing would be required to determine whether these results were statistically significant.

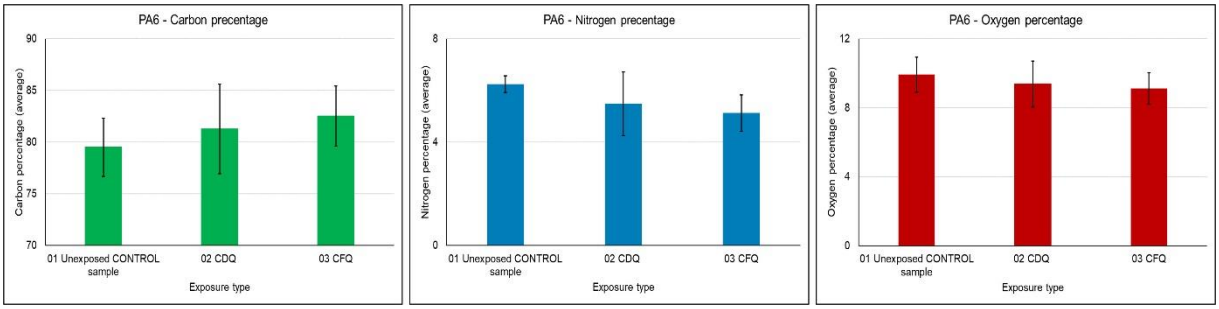

Figure 7: XPS results for the CF-PA6 samples. Carbon percentage is shown on the left, nitrogen percentage is shown in the middle and oxygen percentage is shown on the right. Each graph shows results for the control, CDQ and CFQ cases.

As with the CF-PA6 samples, there was a change in the carbon and oxygen percentage for the CF-PAEK tapes, however the opposite trend was observed. The CDQ and CFQ samples had a lower carbon percentage and higher oxygen percentage, as shown in **Figure 8**. These results are more consistent with the pattern to be expected from oxidative degradation. However, these results could also be explained by an increase in the resin to fibre ratio on the surfaces of the CDQ and CFQ samples. Again, it should be noted that the standard deviation on all these results was large and therefore further testing would be required to draw more confident conclusions.

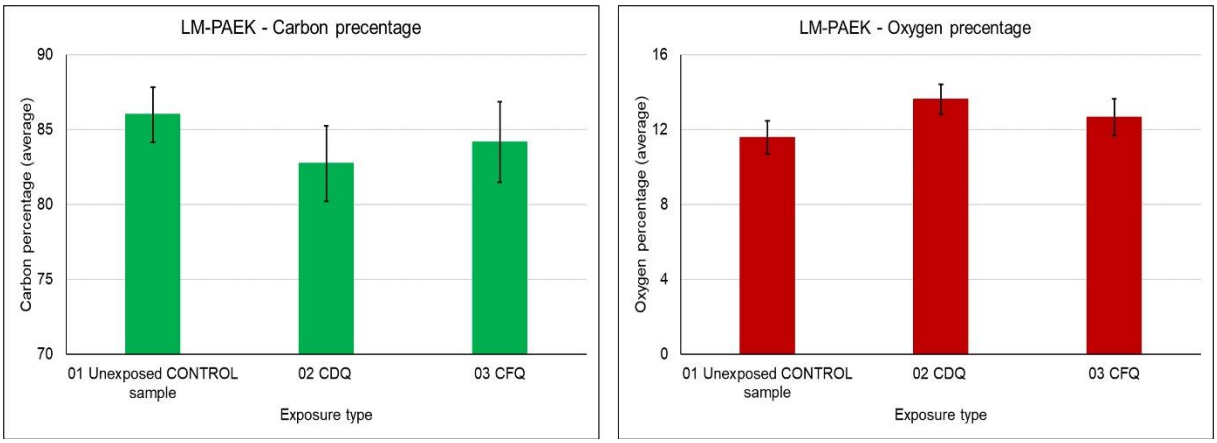

Figure 8: XPS results for the CF-PAEK samples. Carbon percentage is shown on the left and oxygen percentage is shown on the right. Each graph shows results for the control, CDQ and CFQ cases.

Conclusions from Spectroscopy

Three spectroscopy techniques, FTIR, Raman and XPS, were applied to control, CFQ-exposed and CDQ-exposed samples of CF-PA6 and CF-PAEK materials. The complementary tests did not find conclusive evidence that exposure to flash lamp radiation causes chemical degradation of the material surfaces. Of the techniques, XPS showed most promise for further work, possibly combined with similar tests on neat resin to remove the effects of the carbon fibres at the surface.

4 ALTERNATIVE FLASH LAMP TAPE BONDING METHOD

The complexity of the AFP process, with its many interacting parameters, such as compaction force, tape feed accuracy, tool temperature, first ply contact with tool, position of thermal camera and many others make repeatable tests and accurate thermal measurements difficult to achieve. Through reassessing the AFP results on thermoplastic tapes at the NCC, the results of which motivated this project, it became clear that a simpler, more laboratory-scale method of bonding tape pairs would facilitate easier assessment of the effects of single parameters on the bond strength. To this end, a new tape bonding technique was developed, and is described here for the first time, which takes advantage of the flash lamp system's unique ability to apply radiant heat and compaction pressure simultaneously. Under static conditions, it is possible to apply pressure through the quartz light guide at the same time as heating via irradiation through the light guide, creating a precise weld between tapes. The light guide itself does not heat up during the process and is therefore relatively easy to remove from the bonded tapes after processing.

The new method of tape bonding includes the following steps. The previously exposed surfaces of tape pairs are placed in contact, one on top of the other, and covered with a piece of Kapton film, before the quartz light guide is pressed onto the top surface with a known force (**Figure 9**, left). The flash lamp is then used to heat the samples for a given short duration while maintaining the compaction force, to allow the tapes to melt through their thickness and be bonded (welded) together.

In order to bond tape pairs for this project, flash lamp pulse parameters of 200 Volts, 60 Hz, and 1.5 ms, with a heating time of 1 second, were used in all cases. A compressive force of 200 N was applied by tightening a scissor-jack beneath the samples and pressing them onto the light guide of the statically mounted flash lamp head, force being measured on a set of scales beneath (**Figure 9**, right). The compressive force was maintained for 60 seconds after the heating, and a delay of 180 seconds between tests was used to allow the base plate to cool to ambient temperature between tests. Using this method, a 55mm long central section of the tape pairs was bonded (the longer dimension of the rectangular output face of the light guide). A separate project has assessed the ability of this method to bond (weld) multiple layers of thermoplastic material in one short-duration heating step, and results will be published in subsequent papers.

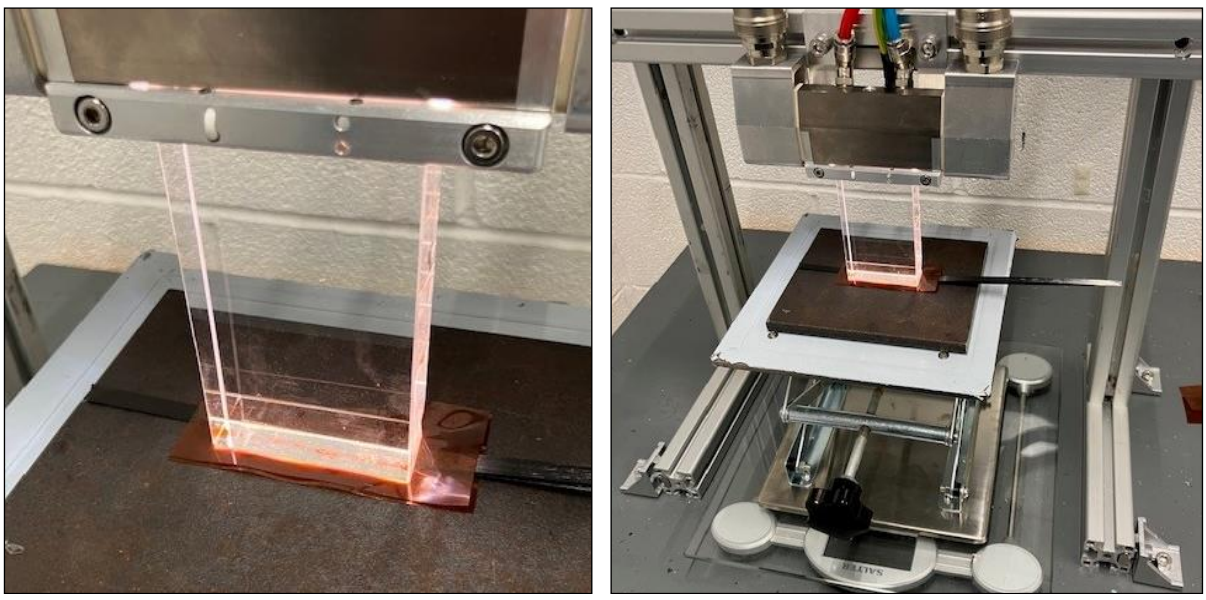

Figure 9: Xenon flash lamp system welding two CF-PAEK tapes under the quartz light guide. Left image: tape pair and Kapton film under the light guide. Right image: the scissor jack and scales.

Measuring Bond Strength of Tape Pairs

Several methods exist in the literature for measuring bond strength of welded thermoplastic interfaces. Standard tests, such as the lap shear test or double cantilever beam (DCB) test, work on thick, rigid specimens of multiple plies. These tests cannot be applied to the thin, flexible tape pairs being considered in this project. Alternatively, several different types of peel testing have been used to characterise the bonding of flexible tapes. T-peel tests, wedge peel tests and floating roller peel tests have been used in the literature ([7], [8]), however T-peel tests are prone to snapping of the tapes prior to interface failure when assessing strongly bonded samples, and wedge peel test results are complicated by the friction between the tapes and the wedge itself. The floating roller peel test ([9]), used throughout this project, provides direct measurement of the bond strength between two flexible tapes along the entire bond surface.

Figure 10 shows the floating roller peel test configuration.

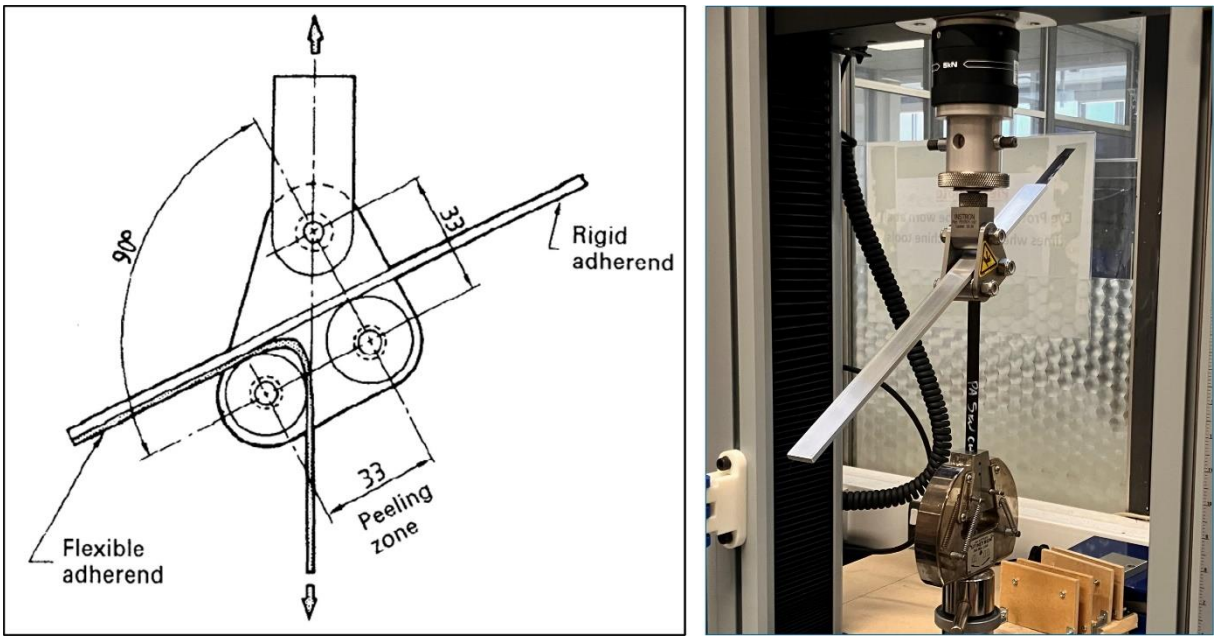

Figure 10: Floating roller peel test configuration. Left: schematic diagram from ASTM D3167−10. Right: picture of floating roller peel test in progress.

Bond Strength Results

Bonded tape pairs from the CDQ-exposed and the CFQ-exposed CF-PAEK material samples were compared with control samples that had not been exposed to flash lamp energy. A total of 10 bonded pairs were created for each category, and bond strengths were measured with the floating roller peel test.

Each tape pair was adhered to an aluminium metal backer using a strip of 3M double coated tape adhesive, 93015LE. The samples were left for at least 24 hours to allow for sufficient adhesive strength between the tape and the aluminium.

The peel tests were carried out on a Instron 5966 screw driven mechanical testing machine, fitted with a 10 kN load cell. The testing methodology was altered from that specified in D3167-10 as the length of bonded tape was only 55mm. The peel rate used was 150 mm/min.

The average force between 10 mm and 20 mm displacement was calculated for each tape pair, then the average of these values was taken over all the samples in each category to give a single number for peel strength of control, CDQ-exposed and CFQ-exposed tapes.

Figure 11 shows the results of the peel tests. The CDQ and CFQ-exposed tape samples, when bonded with the flash lamp welding method, showed a significant reduction in bond strength. There was an 86% reduction in the average force required to peel the tapes apart between the control and CDQ samples and a 98% reduction between the control and the CFQ samples. In general, these results are similar to results

from previous (unpublished) work where AFP was used to bond the tapes. Note that standard deviations, shown with vertical black bars, are high in each category, even though testing was repeated with care.

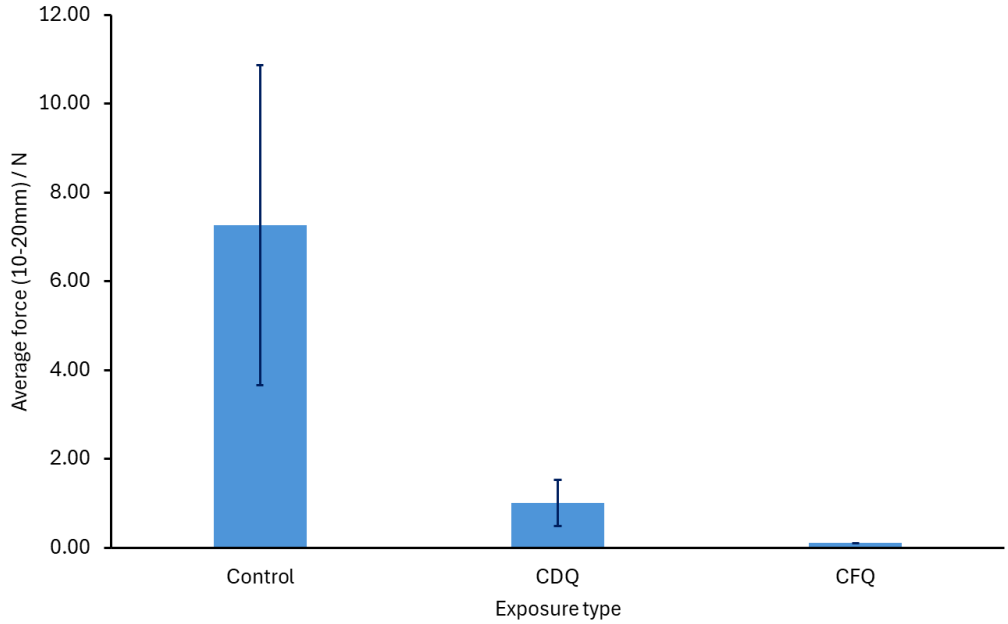

Figure 11: Peel test results from CF-PAEK control, CDQ-exposed, and CFQ-exposed tape pairs

Scanning Electron Microscopy of Peeled Surfaces

Scanning electron microscopy (SEM) images were taken of the peeled surfaces of representative tape pairs to investigate the cause of the different bond strengths seen in the peel tests described in the previous section. Images were taken on a Zeiss Evo MA25 scanning electron microscope. Tape samples were adhered to a 25mm stub using a carbon adhesive tab. The samples were positioned so that the two opposing faces of the bonded pair were next to each other with the peeled surfaces uppermost. The tape samples were coated with a 15nm layer of gold using a Quorum Q150T sputter coater. **Figure 12** shows SEMs of representative peeled surfaces for control, CDQ-exposed and CFQ-exposed samples.

The main observable difference between the control samples, which exhibited high peel strengths, and the CFQ-exposed samples, which exhibited low strengths, was the number of discrete regions of polymer that have failed in a ductile manner, seen in the top images of **Figure 12** as light-coloured ribbons of polymer projecting from the surface, but absent from the bottom images for CFQ-exposed samples. The CDQ-exposed samples exhibited some regions of ductile failure, as shown by the smaller number of ribbons of polymer in the middle image. This type of ductile failure was noted by Comer et. al. ([2]) in their study of PEEK polymer samples and attributed to plastic failure of regions of low crystallinity. The difference between low and high peel strengths was observed to be related to the number of discrete areas of polymer that undergo plastic deformation during the failure of the bond surface.

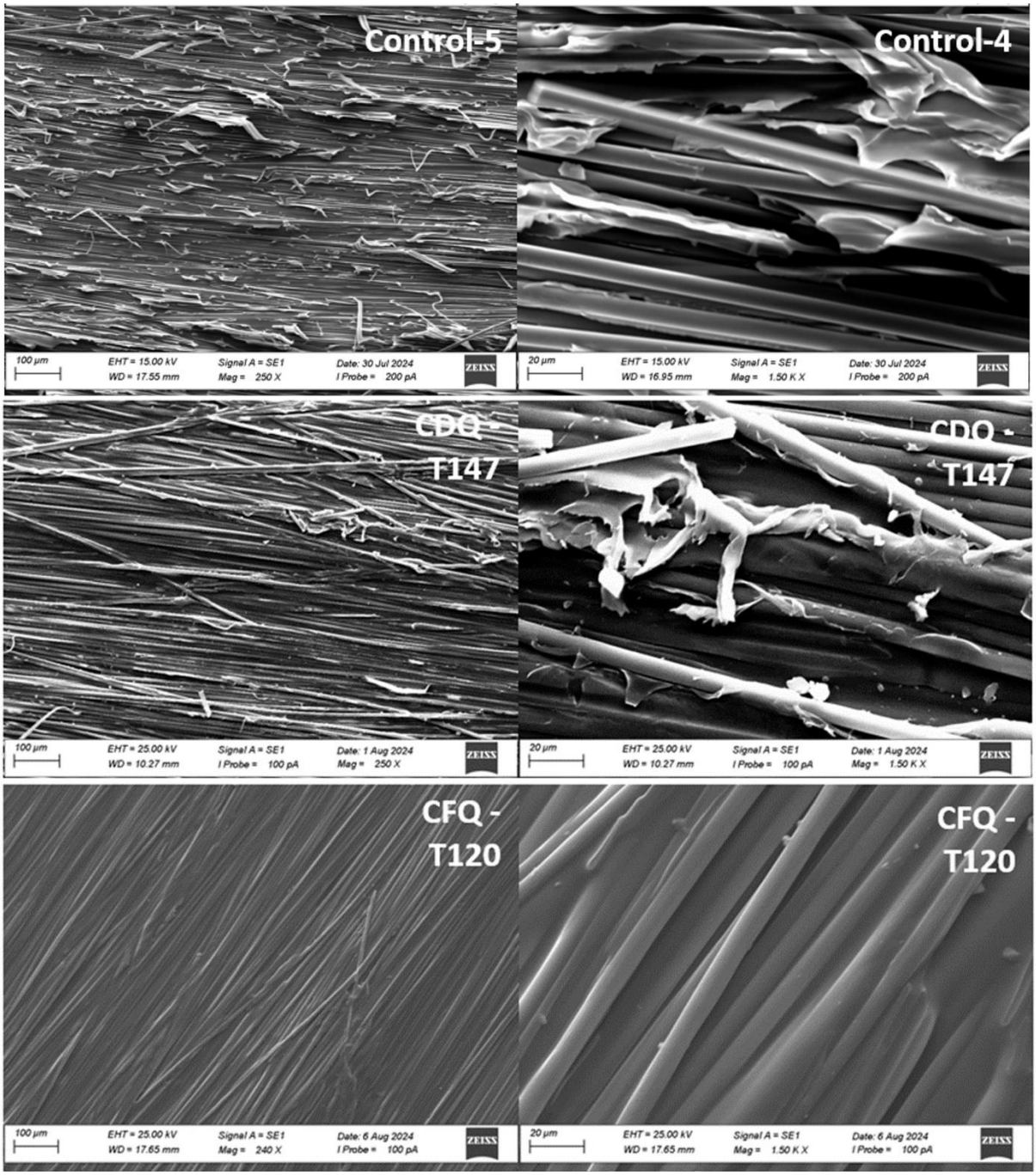

Figure 12: SEM images of peeled tape surfaces for control samples (top), CDQ-exposed samples (middle) and CFQ-exposed samples (bottom). Magnification is 250x (left) and 1500x (right).

Conclusions from Flash Lamp Bonding

Flash lamp bonding of tape pairs from control, CDQ-exposed and CFQ-exposed samples confirmed that exposure to flash lamp energy reduces bond strength. The spectroscopy analysis described in the previous section shows no clear evidence that the polymer surfaces are being chemically degraded by this short duration exposure to the flash lamp energy. An alternative explanation for this reduction in bond strength will form the subject of further work in this area.

5 DISCUSSION AND FURTHER WORK

In this project, no evidence was found that exposure to flash lamp energy caused chemical degradation to CF-PA6 and CF-PAEK tape samples. Further work will be carried out to establish which other factors, present during AFP processing, may detrimentally impact the bond strength between tapes. From the results described here, it appears that reduction in bond strength may not be caused by polymer aging due to UV exposure but by other factors associated with flash lamp heating.

ACKNOWLEDGEMENTS

The main body of this work was completed while the author was a consulting engineer at Excelitas Noblelight (formerly Heraeus Noblelight), Cambridge, UK.

The author wishes to express his gratitude to Martin Brown, Ross Kitson and colleagues at Excelitas Noblelight for supporting this work with funding, technical advice and access to the xenon flash lamp heating system.

The majority of the results shown here were generated through a project at the National Composites Centre funded by Excelitas Noblelight. The author acknowledges with thanks the important contributions of Tom Andrews and Keith Bean at the NCC to the project.

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