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Improved fatigue delamination behaviour of composite laminates with electrospun thermoplastic nanofibrous interleaves using the Central Cut-Ply method

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Abstract

Adding toughening particles to composite laminates is a common approach to increase their delamination resistance. More recently, interleaving the laminated structures with electrospun (thermoplastic) nanofibrous veils is shown to be a viable toughening method. Where toughening composite laminates with nanofibrous interleaves becomes more and more evident under static conditions, the effectiveness under fatigue loadings has yet to be proven. This article provides insight in the nanofibre toughening mechanisms acting under fatigue conditions. Several nanofibre types with a high potential for toughening are considered. A substantial decrease of the delamination propagation rate up to one order of magnitude was obtained for all tested nanofibre types. Furthermore, two distinct zones of delamination behaviour are observed in nanofibre interleaved laminates on exposure to cyclic loading. These insights reveal the crucial design parameters which allow for the production of nanofibre toughened composites with an improved fatigue life.

Keywords: A. Nanocomposites, B. Damage tolerance, B. Fatigue, E. Electrospinning
1. Introduction
The relatively low delamination resistance of most composite laminates remains one of the limiting factors during their in-service life. Increasing the delamination resistance of these materials results in higher damage resistance and improved performance and lifetime. Hence, many methods of interlaminar toughening have been proposed and researched over the past decades [1–12]. The more recent principle of interleaving composite laminates with electrospun nanofibrous veils has shown to be a viable interlaminar toughening method [13–20]. There are several advantages associated to these veils which adds to their commercial applicability. During electrospinning, nanofibrous veils are formed as a self-supporting non-woven membrane which can be handled in a similar way as regular fabrics. They can also be deposited directly on fabrics by guiding dry fabric through an electrospinning set-up [20]. Hence, the veils can easily be placed in between the primary reinforcing plies either as standalone membranes or as nanofibre deposited fabrics prior to composite production and there is no change in the composite manufacturing process required. The nanoscale diameter of the nanofibres offers relatively thin interlayers, while their macroscopic length (continuous fibres) poses no health hazards in comparison with other nanomaterials. The electrospinning process itself is relatively simple in design and proven to be upscalable [22,23] making it a cost-effective nanofibre production method. Furthermore, the electrospinning process used to obtain nanofibrous veils is applicable to many polymer types adding to its versatility [21].

Notwithstanding the numerous expected benefits, the research into composite laminates toughened by electrospun nanofibrous interleaves is still limited. It consists mainly of Mode I and Mode II interlaminar fracture tests performed on interleaved laminates
under static conditions [14,20,24–30]. Although these experiments provide useful information about critical parameters, toughening (micro)mechanisms and general concerns of the delamination behaviour in nanofibre interleaved laminates under static conditions, an analysis of the effect of nanofibrous interleaves under fatigue loadings has not been reported in literature to date. Nevertheless, a thorough understanding of the fatigue delamination behaviour of interleaved laminates is of major concern since almost all composite laminate structures experience fatigue loading during their in-service life. Furthermore, it is known that the delamination behaviour in (toughened) composite laminates can differ substantially between static and fatigue loading [31–33]. This article gives thorough insight into the fatigue delamination behaviour of electrospun nanofibre interleaved composite laminates. Nanofibrous veils are electrospun from polycaprolactone (PCL), polyamide 6 (PA 6) and polyamide 6.9 (PA 6.9) since these systems have proven their effectiveness under static loading conditions [14,27,34]. The focus is on Mode II delamination behaviour as delamination growth during service often occurs under Mode II dominated loadings. The Central Cut-Ply (CCP) method [35] is used to determine the crack propagation rate under cyclic loading as this method is representative for delamination initiation encountered in structural applications, where, for example, delaminations initiate from terminated plies, near free edges or in tapered sections. The observed toughening micromechanisms are linked to the results obtained from static experiments, thus providing the necessary insights in order to design novel fatigue resistant structures.
2. Materials and methods

2.1. Materials

PCL (M_N 80 000) and PA 6 (M_W 51 000 g/mol) pellets were purchased from Sigma-Aldrich; PA 6.9 pellets (M_W 58 000 g/mol) were obtained from Scientific Polymer Products. The polymers are dissolved for electrospinning in a mixture of 98 – 100% purity formic acid (FA) and 98% purity acetic acid (AA), both purchased from Sigma-Aldrich and used as received. Uniform electrospun nanofibrous veils are produced on an in-house developed multinozzle electrospinning machine; more details can be found in Ref. [27]. Standalone nanofibrous veils are produced with an areal density in the range of 14 ± 0.5 g/m². The PCL, PA 6 and PA 6.9 nanofibres had diameters of 650 ± 150 nm, 195 ± 35 nm and 250 ± 30 nm respectively as measured from SEM images (Figure 1a, 1b, 1c).

2.2. Methods

The Central Cut-Ply (CCP) method is used to determine the crack propagation rate under cyclic loading [35]. This method has several advantages compared to the traditional End Notched Flexure (ENF) specimen design such as the use of a simple grip fixture and easy in-situ crack growth monitoring using a clip-on extensometer. Delamination growth in the CCP specimen design initiates from (artificially) interrupted plies instead of an initiation film. Hence, it is much more representative for the type of delamination initiation encountered in structural applications where delaminations initiate for example from terminated plies, near free edges or in tapered sections. The CCP specimen consists of a unidirectional laminated specimen in which a certain number of plies is artificially interrupted symmetrically with respect to the laminate midplane, see Figure 2. When subjected to a far field axial tensile load, four
delaminations will grow under Mode II conditions as interlaminar compression prevents Mode I opening of the delaminations [35].

The Mode II energy release rate (ERR) $G_{II}$ associated with a single delamination can be determined using beam theory as [35]

$$G_{II} = \frac{P^2}{4EB^2t} \left( \frac{\chi}{1 - \chi} \right)$$

with load $P$, Young’s modulus of the unidirectional material without cut plies $E$, specimen width $B$, specimen thickness $t$ and the ratio of cut plies to the total number of plies in the specimen $\chi$ [35]. Equation (1) shows that the Mode II ERR is independent of delamination length and thus remains constant during fatigue tests at constant maximum load. The delamination growth during the fatigue tests is monitored using a clip-on extensometer which is placed in the middle of the specimen. Furthermore, a videocamera is used to monitor the delamination length visually and validate the extensometer measurements. Let $\epsilon^*$ be the strain corresponding to the peak load $P^*$ in each loading cycle during the fatigue tests, the delamination length of a single delamination can then be expressed as [35]

$$a = \frac{1 - \chi}{\chi} L \left( \frac{EBt}{P^*} \epsilon^* - 1 \right)$$

(2)

Differentiation of Equation (2) results in an expression for the delamination propagation rate which is determined by the slope of the extensometer peak strain measurements during the fatigue tests [35]

$$\frac{da}{dN} = \frac{1 - \chi}{\chi} L \left( \frac{E}{P^*} \right) \frac{d\epsilon^*}{dN}$$

(3)

A more detailed derivation and validation of these equations for the CCP method can be found in Ref. [35].
Composite laminates are produced by vacuum assisted resin transfer moulding (VARTM) with unidirectional E-glass plies with an areal density of 500 g/m² (UDO ES500 manufactured by SGL group) and a high toughness epoxy/amine thermoset resin system (EPIKOTE MGS RIMR135 and EPIKURE MGS RIMH137 supplied by Momentive) typically used for wind turbine blades. More details on the used VARTM process can be found in Ref. [36]. A total amount of 8 plies is used with the two middle plies cut prior to stacking. Special care was taken to produce a straight cut along the whole ply in order to have a sharply defined cut region in the final laminate. The standalone nanofibrous veils, which have the same size as a reinforcing ply (300 x 300 mm²), are interleaved in the two interlaminar regions where delamination growth will occur (Figure 1d). The manufacturer’s recommended curing cycle is used to cure the laminates (24 hours at room temperature followed by a post-cure for 15 hours at 80 °C). The nominal thickness of the final laminates is 3 mm. The glass fibre volume fraction (52 vol%) did not change when nanofibrous veils were interleaved as a two-piece mould with a fixed thickness was used to produce the laminates. Furthermore, the nanofibres had no measurable influence on the infusion process and the final quality of the laminates. The infusion resin could easily impregnate the nanofibrous veils in the interlayers. Visual inspection and microscopic images of the interlayers showed no dry spots in the final laminates (transparent epoxy resin) indicating that the porosity did not increase.

Six different laminated plates were produced: two non-interleaved (virgin), two PA 6.9 interleaved, one PA 6 interleaved and one PCL interleaved laminated plate. To validate the CCP results, the same nanofibrous veils were also used to produce composite laminated plates with an initiation film and nanofibrous veil in the midplane for ENF
experiments. The CCP and ENF specimens were cut from the respective laminates with a water-cooled diamond cutting machine to nominal dimensions of 140 x 15 mm² and 140 x 20 mm² respectively. The edges of the specimens were polished in order to remove any edge defects introduced by the cutting machine.

The CCP specimens were tested on an Instron 8801 servo-hydraulic machine equipped with an Instron Dynacell load cell of 100 kN both under static and fatigue loading conditions. Hydraulic wedge grips were used to fix the specimens and the alignment was assured using an Instron alignment kit. An Instron clip-on dynamic extensometer with 25 mm gauge length was used to monitor delamination growth during fatigue tests and mounted centrally on the specimen. Tension-tension fatigue loading was applied at a stress ratio $R$ of 0.1 and test frequency of 5 Hz (load-controlled). Depending on the desired load level, the maximum fatigue stress was adjusted between 260 MPa and 620 MPa. Self-heating of the specimens was monitored using a FLIR T420 infrared camera and was found to be acceptable (maximum heating up to 30 °C at the highest load levels). The equations given in Section 2 were used to analyse the data from the experiments. A Young’s modulus of 42 GPa was used in Equations (1)-(3) as measured on unidirectional samples without cut plies. Quasi-static delamination tests were also performed using the same set-up with a displacement-controlled movement of the grips of 2 mm/min. The first significant drop in load represents the point of major delamination and its value is used to determine the static Mode II interlaminar fracture toughness $G_{IIc,CCP}$ obtained by the CCP method.

The ENF specimens were statically tested on an Instron 3369 electromechanical machine using a load cell of 2 kN. The Beam Theory including Bending Rotations (BTBR) described in Ref. [37] was used to determine $G_{IIc,ENF}$. This method is similar to
the Compliance Based Beam Method (CBBM) used in previous work [14,27], but
corrects for large displacements. The loading roller was displaced at a speed of
1 mm/min. The span length was 100 mm and the ratio of initial delamination length to
half-span length was 0.7 in order to have stable delamination growth.

3. Results and discussion

3.1. Applicability and validation of the CCP method

3.1.1. Applicability to nanofibre interleaved composite laminates
Static CCP tests were performed in order to check if the increase in $G_{IIc}$ for nanofibre
interleaved laminates is not dependent on the testing method and is comparable to the
results obtained in the more commonly used ENF method, i.e. $G_{IIc,CCP}$ should be similar
to $G_{IIc,ENF}$. Cui et al. [38] showed that through the thickness normal compressive stress
is present at the crack tips in CCP specimens. Such compressive stresses are known to
increase the measured Mode II interlaminar fracture toughness [39–41]. In order to
account for this effect, Cui et al. [38] proposed following correction for a glass
fibre/epoxy CCP specimen with a ratio of 2/8 cut plies:

$$G'_{IIc,CCP} = \frac{G_{IIc,CCP}}{1 + 0.008 \times 37.72 \times 10^{-3} \sigma_c} \quad (4)$$

in which $\sigma_c$ is the experimentally determined nominal failure stress of the CCP
specimen. For the CCP specimens reported in this work, correcting for the interlaminar
compressive stress yielded values about 20% lower than directly computed from
Eq. (1). The results show good agreement between the $G_{IIc}$ values obtained in both tests
as shown in Figure 3. The most notable difference between $G_{IIc,CCP}$ and $G_{IIc,ENF}$ is
noted for the virgin laminates with a difference of approximately 15%. This is in
agreement with the difference between both methods reported by other researchers and
is probably related to the simple beam theory used to derive Equation 1 [35].

Furthermore, the load-displacement curves of CCP specimens show a more pronounced stick-slip crack growth (sudden drops in load upon crack extension) compared to the stable crack growth obtained in the ENF tests. This indicates some stress build-up at the crack tip in CCP specimens, which might be due to local through thickness compressive stresses causing a local enhancement in $G_{IIc}$, resulting in an overestimation of $G_{IIc}$.

Although the Mode II interlaminar fracture toughness of the virgin laminates is already relatively high, the nanofibrous interleaves result in a significant improvement up to a toughness of approximately 3000 J/m² which is in the range of thermoplastic composites [42]. Previous research by the authors has shown that this increase is due to the development of so-called nanofibre bridging zones which occur when the delamination path crosses the toughened interlaminar region [27]. Microscopic images of tested CCP specimens show the same delamination crack path behaviour as observed in ENF specimens: regular crossings of the interlaminar region in which nanofibre bridging zones can develop (Figure 4). These crossings are important as they are suggested to be the main cause of an increased interlaminar fracture toughness [43]. In the region between the crossings, delamination progresses at the reinforcing fibre/matrix interface (Figure 4), similar to non-interleaved (untoughened) laminates, and as such is less affected by the electrospun nanofibres. Hence, the same nanofibre bridging toughening mechanism occurs both in CCP and ENF specimens. Furthermore, delamination initiation in a CCP specimen is more “natural” as it does not require an initiation film, and is thus more closely related to the type of initiation that can be expected in composite applications. This is advantageous for testing nanofibre interleaved laminates since we have previously shown that the relative position of the
initiation film and the nanofibrous veil can artificially affect the observed interlaminar fracture toughness resulting in IFT values that may not well represent the actual material [27].

3.1.2. Validation of delamination growth measured by extensometer
As shown above, the CCP method is well suited to determine the Mode II interlaminar fracture toughness of nanofibre interleaved laminates. Furthermore, the CCP method allows for an accurate monitoring of the delamination growth during fatigue testing, as needed to determine the fatigue delamination behaviour, from relatively simple extensometer measurements through Equation (3). Figure 5 shows the delamination length - as calculated by Equation (2) from the strain measurements – as a function of the amount of loading cycles for virgin (non-interleaved) laminates at a high and a low value of load severity $G_{II max}/G_{II c, virgin}$. The delaminations grow linearly with the amount of loading cycles and the slope of this curve is equal to the delamination growth rate $da/dN$ as defined by Equation (3). A linear delamination growth behaviour is observed for all virgin specimens, independent of the applied load severity. In addition, a visual observation of the delamination front also enables the calculation of the delamination growth rate by measuring the delamination length after a certain amount of loading cycles. Figure 6a reveals that the delamination growth rate determined by Equation (3) agrees well with the growth rate determined from visual observation. Hence, the CCP method provides the means to accurately determine the delamination length in-situ based on relatively simple strain measurements. This is advantageous as the Mode II delamination front is usually difficult to determine visually from the edges of a specimen when the laminates are not translucent [44]. The delamination growth rate
of the virgin laminates decreases proportionally to the load ratio and the datapoints fit well to the semi-empirical model proposed by Allegri et al.

\[
\frac{da}{dN} = C \left( \frac{G_{ll,max}}{G_{llc,virgin}} \right)^b \left(1-R\right)^2
\]  

(5)

where \(C\) and \(b\) are empirically derived constants [35], see Figure 6b. The datapoints in Figure 6b were fitted to Equation (5) using a least squares regression algorithm. The model gives a linear dependency between \(\frac{da}{dN}\) and \(\frac{G_{ll,max}}{G_{llc,virgin}}\) on a double logarithmic scale. Table 1 represents the fitting coefficients and a quantitative measure for the fitting accuracy of Equation (5) using the coefficient of determination (CoD).

The semi-empirical model describes the behaviour of the virgin laminates well with a CoD of 96.4%.

3.2. Analysis of the fatigue behaviour of nanofibre interleaved composites

3.2.1. Delamination growth behaviour

The delamination growth behaviour during the fatigue testing of nanofibre interleaved specimens is found to be different from that of the virgin specimens. A distinct transition in delamination propagation rate was often observed after a few millimetres of delamination growth in the CCP specimens. Initially, the delamination grows relatively slowly for several millimetres at a constant rate after which the delamination growth rate increases to values similar to those of the virgin specimens. This is schematically represented in Figure 7. Microscopic images of the cross-section of failed CCP specimens taken near the initiation region showed the same delamination behaviour as observed in the static experiments, i.e. regular crossings of the nanofibre toughened interlaminar region (Figure 4). At several millimetres away from the initiation region, some specimens showed almost complete glass fibre/epoxy debonding with none or very few interlaminar crossings. Hence, at these points, the delamination growth rate
starts to approach that of the virgin non-interleaved specimens as the amount of interlaminar crossings is minimal. Observation of the fracture surface indicates that the distance between two neighbouring interlaminar crossings can become smaller with increasing delamination growth. Eventually, both crossings combine at a certain point of delamination growth after which the delamination grows by glass fibre/epoxy debonding without crossing the interlayer (Figure 8). This mechanism of interlaminar crossing suppression causes the distinct delamination behaviour observed in nanofibre interleaved specimens.

The transition from a delamination path with interlaminar crossings (Region I delamination growth) to one without (Region II delamination growth) is not instantaneous, but spans a certain amount of cycles and a certain amount of delamination growth. This transition zone is associated with the disappearance of individual interlaminar crossings. Observation of the fracture surface showed that these crossings do not disappear at the same time (same point of delamination growth). In addition, the transition zone is further broadened as this effect is present at each of the four delamination interfaces.

3.2.2. Effect of load severity on delamination growth behaviour

The transition from Region I to Region II delamination growth in nanofibre interleaved specimens seems to be predominantly present at low load levels, while a constant delamination growth rate is more often observed at high load levels (Figure 9).

Inspection of the fracture surface of failed specimens showed that the suppression mechanism is still present at high load levels, but takes place after a longer length of delamination. As such, the transition from Region I to Region II delamination growth was not always visible on the delamination growth data as it happened at delamination
lengths higher than the maximum measurable delamination length (using an extensometer of 25 mm travel).

Particularly at the lower load levels, the driving force for the suppression of interlaminar crossings seems to be high, which results in complete glass fibre/epoxy interfacial failure without crossings after several millimetres. As the amount of delamination growth per cycle decreases substantially at lower load severities, the delamination has more time (i.e. more cycles) to realign itself in a more energetic favourable position outside of the toughened interlayer (i.e. at the glass fibre/epoxy interface) after only a few millimetres. Furthermore, the plasticity of the epoxy matrix increases at lower strain rates [45] which adds to the driving force for the suppression of interlaminar crossings at low load severities as the increased plasticity also causes an increased matrix toughness.

The interlaminar crossing suppression was also observed on the fracture surfaces of CCP and ENF specimens tested under static conditions, but there it took several centimetres before all the interlaminar crossings have disappeared. Hence, the mechanism of interlaminar crossing suppression seems to always be present in nanofibre interleaved specimens, but the length at which all the crossings have disappeared depends on the type of loading (high load severity, low load severity, static). Furthermore, it also depends on the type of nanofibre system (Figure 9). In general, for the same kind of loading, PCL nanofibres result in a longer Region I delamination growth compared to PA 6.9 and PA 6 nanofibres.

**3.2.3. Improved fatigue delamination resistance**

The delamination growth rate as a function of the load severity for the nanofibre interleaved laminates is given in Figure 10. The plotted delamination growth rates were
calculated from strain measurements in Region I. All nanofibre interleaved specimens showed an overall decrease in $da/dN$ as compared to the virgin (non-interleaved) specimens, indicating an improved delamination resistance under fatigue loading. Improvements in delamination growth rate up to 15 times compared to the virgin material were obtained for individual specimens. It is worth noting that the PCL nanofibre interleaved specimens performed best over the range of load severities tested. Furthermore, the delamination behaviour of the nanofibre interleaved laminates is well described by the semi-empirical model of Equation (5) with CoD values between 90 - 95% (Table 1). As described in Section 3.1.1., the improved delamination resistance can be associated with the formation of nanofibre bridging zones in interlaminar crossings. Analysis of the fracture surface of tested CCP specimens showed that these nanofibre bridging zones also form under fatigue loading conditions, see Figure 11. They mainly develop at interlaminar crossings where the delamination causes a relatively large zone of interlaminar failure, but also occur at other interlaminar (micro)cracks, for example during hackle formation (tensile microcracking at 45° relative to the delamination plane).

A more detailed SEM analysis of the nanofibre bridging zones in interlaminar crossings is represented in Figure 12. The electrospun nanofibres protrude from the epoxy resin and show a high degree of plastic deformation (Figure 12a and Figure 12c). Their morphology is typical for static tensile failure of (thermoplastic) fibres with a clearly defined necked region resulting in a tip-like fibre end. A similar nanofibre morphology is observed on statically tested CCP specimens. Crazing is observed at the interface between PCL nanofibres and epoxy indicative of a good adhesion between both polymers (Figure 12b) as reported earlier [43]. In the case of polyamide nanofibres, the
interface is mainly governed by relatively weak Van der Waals forces or hydrogen bonds resulting in a relatively weak PA-epoxy interface [46]. Indeed, the PA nanofibres occasionally debond from the epoxy resin without much deformation, resulting in smooth nanofibre imprints in the epoxy (Figure 12d). This peeling mechanism most likely results in a lower energy uptake than if the nanofibres would plastically deform. Hence, the difference in interface quality between PCL-epoxy and PA-epoxy can explain why PCL nanofibres outperform the PA nanofibres in the fatigue tests.

**Conclusion**

This article describes the Mode II delamination behaviour of composite laminates toughened with electrospun nanofibrous interleaves under fatigue loading. The Central Cut-Ply method is used as it allows for a natural crack initiation into the nanofibre toughened interlayers similar to the initiation mechanism expected during service. The CCP method is found to give similar results for the Mode II interlaminar fracture toughness as the more commonly used ENF method. The CCP method however allows for an accurate monitoring of the delamination growth during fatigue testing from relatively simple extensometer measurements. This was validated by comparing the delamination growth rate obtained by strain measurements with the actual delamination growth rate obtained from visual observation of the delamination front.

Static experiments showed that the increase in interlaminar fracture toughness is associated with the development of nanofibre bridging zones at interlaminar crossings. Under fatigue loading, the same mechanism of nanofibre bridging at interlaminar crossings is observed. This results in a substantial decrease in delamination growth rate up to one order of magnitude, and thus, an improved delamination behaviour under fatigue conditions of nanofibre interleaved composite laminates. A distinct fatigue
delamination behaviour is observed for nanofibre interleaved laminates: depending on the applied load severity, there is a tendency for suppression of interlaminar crossings after a certain amount of delamination growth. The delamination growth of nanofibre interleaved specimens thus exhibits three regions during fatigue testing. Initially (Region I), the delamination propagation rate remains relatively constant and is smaller than the rate of the non-interleaved material. As more and more interlaminar crossings disappear, the delamination growth rapidly increases and the delamination growth rate of the nanofibre interleaved specimens approaches the rate of the non-interleaved material when no interlaminar crossings remain (Region II). Both regions are separated by a transition zone in which the interlaminar crossings combine and disappear. Especially at low load severities, the driving force for this suppression mechanism is found to be high.

All three nanofibre types tested, i.e. PA 6, PA 6.9 and PCL, result in improved fatigue delamination resistance, and the PCL interleaved laminates have the best performance over the range of load severities tested. Analysis of the fracture surfaces with SEM shows bridging nanofibres protruding from the epoxy matrix. Upon crack extension, these nanofibres will strain, yield and fail resulting in a significant amount of absorbed energy. Despite the fact that the CCP specimens are subjected to fatigue loading, the protruding nanofibres failure mode is similar to that found for (quasi-)static tensile failure of polymer fibres with a necked region resulting in a tip-like fibre end after failure.

The results obtained in this work indicate that nanofibrous veils are suitable as an interlaminar toughener under both static and fatigue (Mode II) loading conditions as they improve the delamination resistance considerably. Furthermore, the veils are easily
integrated in composite laminates without any changes to the composite production process or the quality of the final laminates. As such, they are a viable interlaminar toughening material for high-end and demanding composite applications.

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5. References


Figure 1: SEM images of electrospun PCL (a), PA 6 (b) and PA 6.9 (c) nanofibres. The nanofibrous veils are subsequently interleaved at the delamination interfaces near the cut-ply region (d).
**Figure 2:** Central cut-ply specimen design. Four delaminations emanate under Mode II loading conditions from the interrupted plies due to far field axial tensile loading. The delamination growth is monitored in-situ by an extensometer and a videocamera.
Figure 3: Nominal axial stress versus displacement curves for representative CCP specimens show that $G_{IIC}$ increases upon interleaving the laminates with nanofibrous veils (a). Similar results are obtained using the standard ENF method (b) which indicates that the applicability of the CCP method to determine the $G_{IIC}$ of nanofibre interleaved laminates is good.
Figure 4: The delamination path in an interleaved CCP specimen shows interlaminar crossings through the nanofibre modified interlayer. Nanofibre bridging zones mainly develop in these crossings resulting in an improved toughness.
Figure 5: Delamination length in function of the amount of loading cycles at low (a) and high (b) load severity. The delamination grows linearly with the amount of loading cycles and the slope corresponds to the delamination growth rate.

Figure 6: Delamination growth rate $da/dN$ in function of the applied load for virgin (non-interleaved) specimens (each point represents an individual specimen). Good agreement is observed between $da/dN$ determined by strain measurements according to Equation (3) and $da/dN$ determined by visual observation of the delamination front (a). The virgin material's fatigue behaviour is well approximated by the semi-empirical model of Equation (5) (b).
Figure 7: The delamination growth of nanofibre interleaved CCP specimens exhibits three regions during fatigue testing: (i) relatively constant delamination growth rate smaller than that of the non-interleaved material (Region I), (ii) rapidly increasing delamination growth rate due to suppression of interlaminar crossings (transition zone), and (iii) delamination growth rate similar to that of the non-interleaved material (Region II).
**Figure 8:** Interlaminar crossings get suppressed, resulting in complete interfacial debonding without interlaminar crossings after a certain distance of delamination growth. Schematic representation of cross-section views (a). Fracture surface of a failed CCP specimen with two interlaminar crossings that disappear after 2 – 3 mm of delamination growth (b).

\[ \frac{G_{\text{ILmax}}}{G_{\text{ILc,virgin}}} = 0.15 \text{ (low)} \]

\[ \frac{G_{\text{ILmax}}}{G_{\text{ILc,virgin}}} = 0.45 \text{ (high)} \]

(a)  

**Figure 9:** At low load levels, nanofibre interleaved specimens show a substantial reduction in delamination growth rate compared to the virgin specimens during Region I delamination growth (a). At higher load levels, a constant delamination growth rate is more often observed throughout the test (b).
Figure 10: The delamination growth rate decreases for laminates interleaved with PA 6.9 (a), PA 6 (b) and PCL (c) nanofibrous veils (each point represents an individual specimen). Overall improvements up to one order of magnitude are obtained for the nanofibre interleaved laminates. The PCL interleaved specimens show the best improvements over the whole load severity range considered (c).
Figure 11: Bridging nanofibres are observed on the fracture surface of tested CCP specimens at interlaminar crossings (a), in interlaminar microcracks (b), and at the base of hackles (c).
Figure 12: Detailed SEM images of bridging nanofibres observed on interlaminar crossings: protruding PCL nanofibres (a), crazing observed at the PCL-epoxy interface (b), protruding PA 6.9 nanofibres (c), and imprints left by debonded PA 6.9 nanofibres (d).
Table 1: Fitting parameters $C$ and $b$ for Equation (5) and the resulting fitting accuracy determined by the coefficient of determination (CoD).

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<tr>
<th>Specimen series</th>
<th>$C$ (mm/cycle)</th>
<th>$b$</th>
<th>CoD (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Virgin (non-interleaved)</td>
<td>0.65</td>
<td>3.18</td>
<td>96.9</td>
</tr>
<tr>
<td>PA 6.9 interleaved</td>
<td>0.08</td>
<td>2.95</td>
<td>94.1</td>
</tr>
<tr>
<td>PA 6 interleaved</td>
<td>0.10</td>
<td>2.68</td>
<td>94.3</td>
</tr>
<tr>
<td>PCL interleaved</td>
<td>0.10</td>
<td>3.28</td>
<td>90.3</td>
</tr>
</tbody>
</table>