THERMAL AND MECHANICAL CYCLING OF THIN-PLY COMPOSITES FOR CRYOGENIC APPLICATIONS

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Abstract

Currently, huge undertakings to develop concepts for fossil free aviation are being made. For instance, hydrogen gas can be used in fuel cells generating electricity for motors or in fossil free combustion engines. To minimize the volume, the hydrogen must be stored in liquid form in tanks at very low temperature (-253°C). These tanks should preferably have as low weight as possible, which may be obtained by using carbon fiber reinforced polymer composites. However, pressure and temperature changes during fueling can cause microcracks between the fibers, which then causes gas leakage. By using thin composite plies of different orientations, the formation of microcracks can be suppressed. However, the damage development due to cryogenic cycling and its effect on long term performance is not well understood. This work aims at reducing this knowledge gap by characterizing thin ply composites under cryogenic thermo-mechanical fatigue. In this work, the materials (carbon fiber and matrix) were selected and cross ply $[90/0]_{4s}$ composite laminates were manufactured using wet filament winding. The laminates were inspected for damage, and samples prepared for testing. Quasi-static, mechanical fatigue and thermal fatigue tests were performed. Only a few matrix cracks were observed at a very high load and high number of cycles. Those cracks were initiated but not propagated along the width of the specimens. The results show that they have potential for being used in ultralight tanks for liquid hydrogen.

Keywords: Thin ply composites, Transverse cracking, Cryogenic temperature, Fatigue

1. Introduction

Aeronautical and aerospace applications are increasingly using composite materials such as carbon fiber/epoxy laminates, justified by their outstanding combination of properties, including high stiffness, strength, and low density. Despite these advantages, a significant challenge faced by composites in various applications is the occurrence and accumulation of micro-damage during their service life. When subject to increasing tension, a multi-layered composite laminate will eventually experience macroscopic failure. However, the ultimate failure is typically the result of multiple micro-damage mechanisms that initiate and develop throughout the loading history. In polymeric composites, the first mode of micro-damage often involves the formation of transverse (intralaminar) cracks in the off-axis layers. This is followed by the growth of delamination and the development of fiber breaks in adjacent plies around the tips of the transverse cracks, attributed to stress concentrations. The final macroscopic failure of the laminate depends on the sequence and extent of the micro-damage events during the loading history. Numerous studies have been published examining the effect of damage development in composite laminates on thermo-elastic properties [1-5]. Hence, to improve failure resistance and enhance the overall mechanical efficiency, it is desirable to inhibit the formation of micro-damage, especially the onset and progression of transverse cracks, which frequently act as the starting point for subsequent damage mechanisms.

An effective approach to suppress the formation of transverse cracks involves decreasing the thickness of the plies in the laminate. This reduction lowers the potential energy release rate (ERR) for crack propagation along the fiber direction within the ply, thereby demanding a greater applied load to initiate a tunneling crack [6-12].

Lower probability of significant defects in a thin layer can enhance resistance to cracking. Matrix cracking, delamination, and splitting damage occurring under static, fatigue, and impact loadings can be effectively postponed without the necessity for specialized resins or through-the-thickness reinforcements. Numerous experimental studies have substantiated this claim [9-17]. The impact of ply thickness on crack propagation mechanisms was examined in [9], where the energy release rate of intralaminar transverse cracks was investigated through finite element analysis. The investigation verified the crack suppression effect in thin-ply laminates, ultimately attributing this effect to a reduction in the energy release rate at the crack tip in the thin layer [9].

In [14], the strength of quasi-isotropic carbon/epoxy laminates was examined. The findings revealed that in thin-ply quasi-isotropic laminates, the onset of damage can reach as high as 92% of the ultimate fiber failure strain level. In contrast, conventional thickness laminates exhibited damage onset at 41-66% of the ultimate fiber failure strain. The literature contains a huge number of analytical and numerical studies on thin-ply laminates, as represented by [14, 17-21]. A thorough examination of analytical and numerical models for predicting the mechanical response and damage in thin-ply laminates is presented in [19]. Within this context, numerical studies conducted in [19-21] analyzed the effect of ply thickness on the in-situ transverse strength, revealing a significant enhancement in both transverse strength and resistance to delamination in thin-ply laminates. The analytical models for transverse crack propagation, based on fracture mechanics and introduced in [7, 8], have been widely implemented in the analysis of transverse cracking in thin-ply laminates, as proven by studies in [14, 16-21].

Additionally, in [22] analytical solutions and closed-form expressions for the energy release rate during steady-state propagation (tunneling) of non-uniformly distributed transverse cracks in both Mode I and Mode II are derived, highlighting the dependence on ply thickness.

Experimental data concerning the evolution of micro-cracks are typically acquired through observing specimen edges under stress magnification. Strictly speaking, the observed crack density at the specimen edge has a weak connection with crack propagation along fibers within the ply, which is governed by the energy release rate (ERR) and influenced by ply thickness. The crack density at the edge of thin-ply laminates can be significantly higher than the crack density within the bulk of the ply. The statistics of edge cracks explain the initiation and propagation of cracks in the ply thickness direction, which, upon reaching a critical size, becomes unstable.

Crack initiation is a complex phenomenon involving the coalescence of growing fiber/matrix debonds through microcracks in the matrix. The stress required for this initiation varies in different positions within the ply due to local stress concentrations dependent on non-uniform local fiber distribution (clustering).

The examination of the ply stress level for crack initiation and its potential correlation with ply thickness has been seldom studied. Literature suggests that the stress required for crack initiation is largely independent of ply thickness [23-25]. In [25], a computational micromechanics analysis was conducted to investigate crack initiation through debonding (cohesive elements) and matrix yielding, utilizing the fiber distribution in the 90-ply extracted from micrographs. In [24,25], the dependence of fiber/matrix interface crack growth on the proximity of a stiff layer was analyzed, revealing an observable effect only when the debonding fiber is closest to the ply interface.

Thin-ply laminates exhibit promising crack suppression attributes, offering potential benefits in applications such as bolted joints in composite structures under bearing loads. Previous studies, as referenced in [26,27], have explored the use of thin plies to enhance the bearing strength of composite laminates. The results indicate that as the number of thin plies within the stack increases, there is a proportional improvement in material performance such as bearing stiffness, strength at the onset of damage, and ultimate bearing stress.

Another potential application where thin-ply laminates may be used is in cryogenic composite fuel tanks for future aerospace vehicles. These laminates hold the potential to serve as alternatives to current designs of composite fuel tanks, which rely on metallic or polymer-based liners to prevent leakage in pressurized structures. By employing thin-ply laminates with robust crack suppression features, the need for liners could be eliminated, leading to a more lightweight structure, commonly referred to as a Type V tank.

In [28] ultra-thin ply carbon fiber/epoxy cross-ply laminates subjected to tensile loading at room, –50, and –150°C temperatures relevant for cryogenic fuel storage, aeronautical, and aerospace applications were studied. Regarding crack propagation, it was found that in most cases even at very high applied strain levels (1.5%) only a few transverse cracks have propagated from the specimen edges to its middle.

In this work, the effect of thermo-mechanical cycling of thin ply composites intended for cryogenic hydrogen storage applications was investigated. Mechanical quasi-static and fatigue as well as thermal fatigue tests were performed. Only a few matrix cracks were observed at very high load and high number of cycles. Those cracks were initiated but not propagated along the width of the specimens. These results show that thin ply composites have the potential to become viable material solutions for future lightweight hydrogen tanks.

2. Materials and manufacturing Use of this Template

The material in this study consisted of a dry TeXtreme® thin ply unidirectional UD band carbon reinforcement manufactured by Oxeon, which during the winding operation was impregnated by a special epoxy resin suited for cryogenic use. The thin ply UD bands were 20 mm wide, about 0.05 mm thick. Nominally [90/0]4s laminates were manufactured by filament winding of the impregnated bands on a flat 500 mm x 500 mm x 50 mm aluminium tool plate with 25 mm edge radius. The biaxial layup was realized by adjusting the holders at two diagonal corners of the tool plate, Figure 1. The winding was followed by a cure cycle of 24 h in +66°C and 4 h in +93°C. The plates have a thickness of 1.48 mm with a resulting fiber volume fraction was 54.5 %, and a void content of less than 1 %.

Rectangular specimens of 20 mmx200 mm with a gauge length of 100 mm were cut by waterjet. Tensile and mechanical fatigue specimens were equipped with 1.5 mm thick and 50 mm long loading tabs from glass fabric/epoxy in a ±45° orientation, where the end at the gauge section was chamfered at 15° through the thickness.

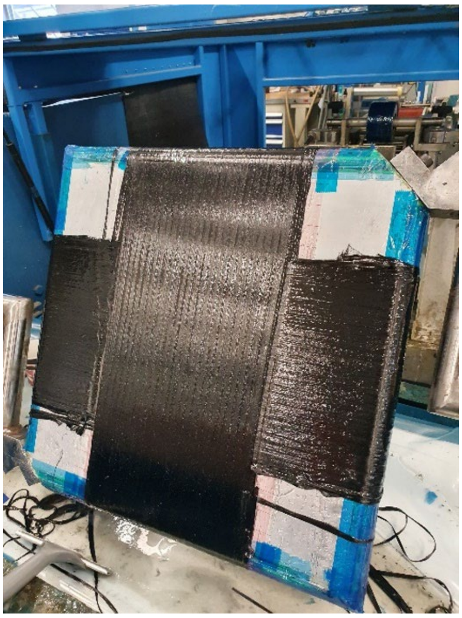

Figure 1 – Nominally bidirectional laminate on winding tool before curing.

3. Experimental results

3.1 Damage evolution in cross-ply laminates due to quasi-static tests

Damage evolution in cross-ply specimens subjected to tensile load was studied at room temperature. All tests were performed using an Instron 5582 testing machine equipped with a 100 kN load cell.

Specimens were loaded and unloaded in steps. In each step a certain selected maximum strain level was reached after which the specimen was unloaded and taken out from the testing machine grips. After each loading step the polished edges of each specimen were studied under an optical microscope and the number of cracks was counted within the gage length of 50 mm in the middle of the specimen. Thereafter, the specimens were mounted back in the testing machine and subjected to the next loading step with a higher maximum strain level (by 0.2% higher than is the previous step). The manual counting of transverse cracks was performed after each load step.

The first investigations were done when no loading was applied during the inspection, however, transverse cracks were not clearly visible. Only fiber breaks were observed at very high applied strain and starting from 1.1% strain as it is shown in Figure 2.

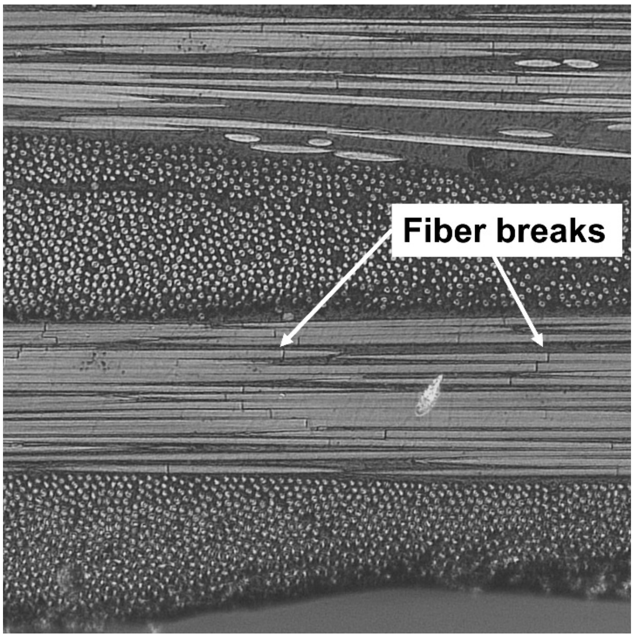

Figure 2 – Fiber breaks at 1.5% strain.

For that reason, we have investigated the following options to open the matrix cracks if they existed:

- Option 1: A small tensile machine (Minimat Tensile Testing Machine) was used while looking in the microscope. The maximum load applied with the small tensile machine was about 500 N which gives an equivalent strain of 0.04% which is considered as very low strain. Only fiber breaks were observed with this method. Figure 3 described how the small tensile machine looks and how it was used.

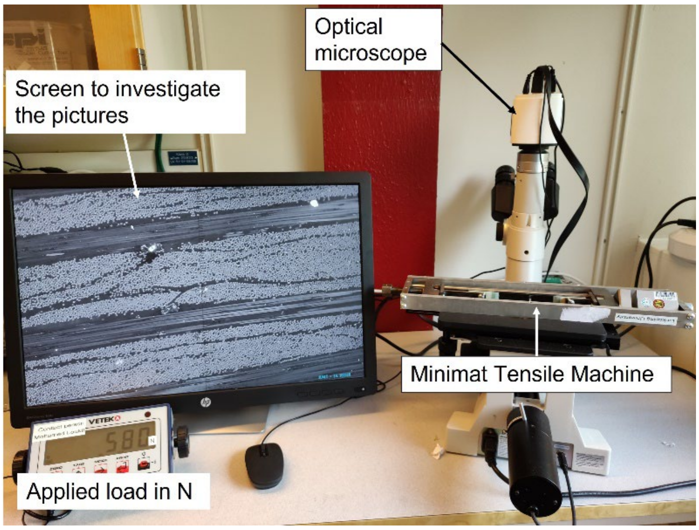

Figure 3 – Specimen loaded during microscopic investigations.

- Option 2: The specimen's edge was coated with white paint while subjected to a load of 900 N (0.07% strain). The process involved applying white paint to the polished edge, allowing it to penetrate the transverse cracks, removing the excess paint, and subsequently examining the edge using an optical microscope. Figure 4 describes the specimen during the painting procedure.

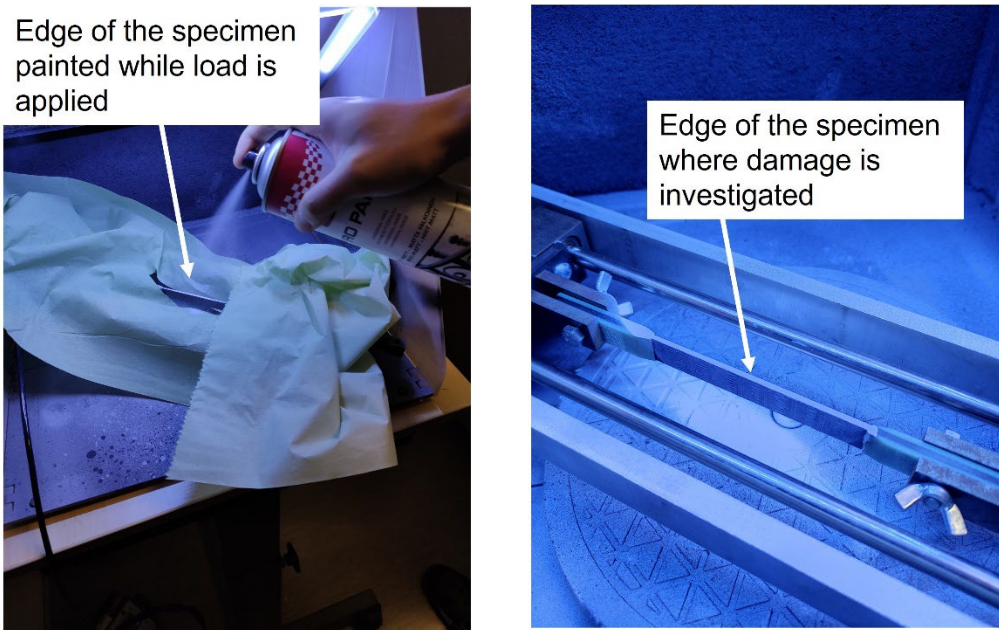

Figure 4 – Paint applied while specimen is loaded.

By employing this approach, only two matrix cracks were detected following the specimen's loading up to 1.5%. These two cracks are illustrated in Figure 5 with enhanced visibility of inspection.

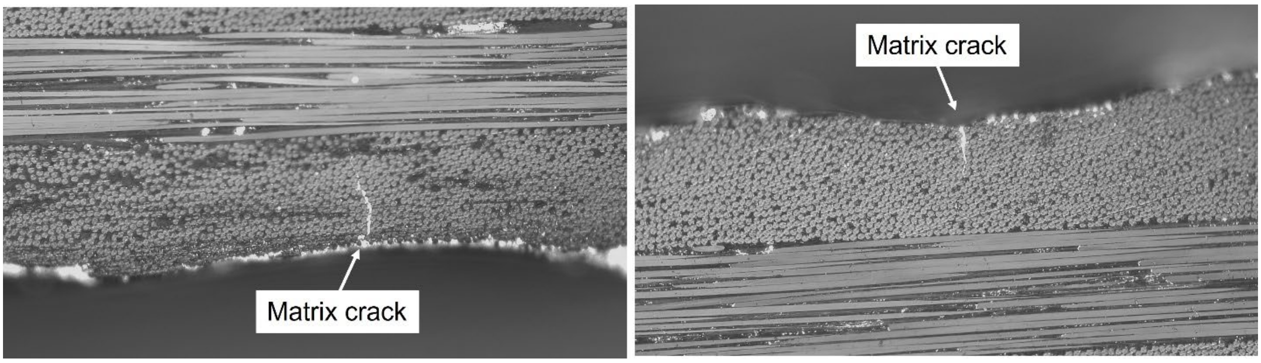

Figure 5 – Two matrix cracks were identified using the paint.

- Option 3: Edge replication. A replica was used to identify cracks while the specimen was loaded to 0.1% of strain. This method involves the microscopic examination of surface replicas prepared by pressing a polymer against the specimen edge. Few cracks were observed using this method. Figure 6 presents a selection of matrix cracks captured at 1.6% applied strain.

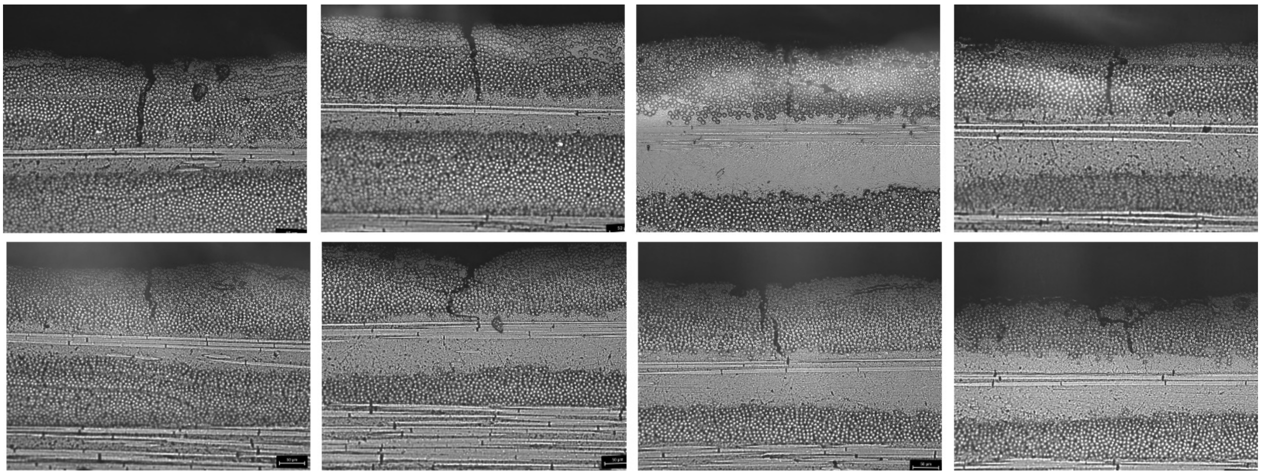

Figure 6 – Transverse cracks observed in the surface 90 layer at 1.6% strain (0.1% was applied to open the cracks). Pictures taken from replica.

The evolving transverse crack initiation in the damaged 90-ply observed at the specimen edge is characterized by the "crack density". The transverse crack density is calculated as the number of initiated transverse cracks per gage length Lg=50mm.

The table provided below (Table 1) summarizes the test results obtained from the evaluation of four specimens. For specimen 1, only option 1 was used, and no transverse cracks were observed until failure. For specimen 2, option 1 and option 2 were employed resulting in the identification of a few transverse cracks. For specimen 3 and 4, option 3 was applied which led to the observation of a greater number of cracks. For all investigated specimens, only cracks on the surface 90 layers were observed. Significantly, fiber breakages became apparent in all specimens at an applied strain of 1.1%. Ultimately, specimen failure occurred within the range of 1.6% to 1.7% of the applied strain.

Table 1 – All results from the quasi-static tests

Specimen	Thickness (mm)	Ex (GPa)	Maximum strain (%)	Number of matrix cracks (length 50 mm)	Fiber breaks
1	1.8	63	1.69 % (failed)	0 (unloaded specimen)	Yes (from 1.1%)
2	1.9	62	1.60% (failed)	4 (with applying a small load 500 N)	Yes (from 1.1%)
3	1.9	58	1.70% (failed)	8 (with replica, 1.6% Strain)	Yes (from 1.1%)
4	1.9	62	1.63% (failed)	12 (with replica, 1.6% strain)	Yes (from 1.1%)

The crack density as a function of applied for is shown in Figure 7. Transverse cracks were identified using option 3, and specimen 4 was thermally shocked 20 cycles (20 seconds at liquid nitrogen, and 10 min at 70 degree) before it was tested mechanically. The intension of using thermal shock was to investigate if the transverse cracks will appear faster than the unshocked specimen.

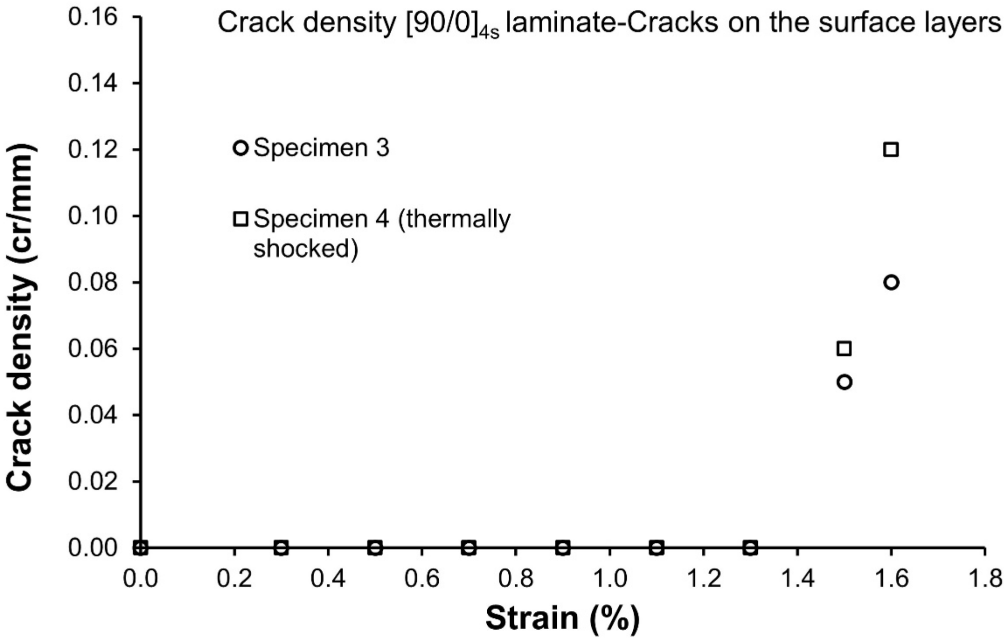

Figure 7 – Crack density as a function of strain (cracks measured with replica).

3.2 Damage evolution in cross-ply laminates due to mechanical fatigue

Fatigue cycles were applied, R=0.1, Frequency 5Hz, cracks were checked at 100, 1000, and 50 000 cycles. Only a few matrix cracks were observed after 50000 cycles. Figure 8 presents some of the matrix cracks. Cracks were observed only on the surface 90 layers.

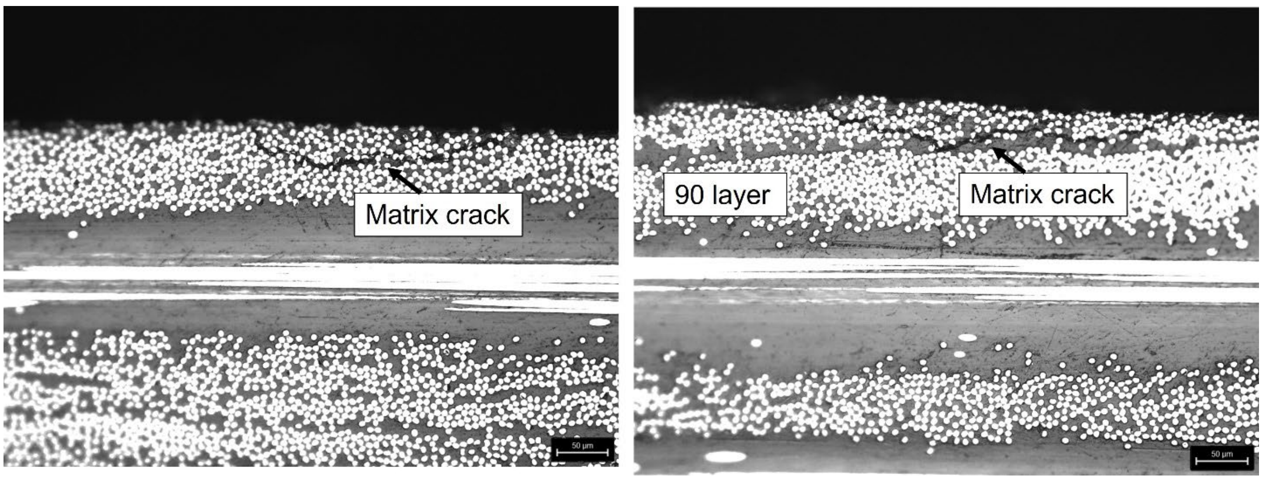

Figure 8 – Matrix cracks after 50000 cycles.

4. Thermal cycling

The possible effect of thermal cycling on the crack initiation at the specimen edge was evaluated in a separate study where mechanical load was not used and only the temperature was cycled. Cross-ply laminate specimens from the same plate used in the mechanical tests were subjected to cyclic thermal loading by submerging specimens in liquid nitrogen (temperature -196°C), and then +70 °C environment for 10 minutes. The higher temperature was chosen to expose the specimens to a higher ΔT. For handling purposes, the specimen ends were attached to metallic wire through pre-drilled holes. The described set-up allowed simultaneous cycling of multiple specimens and allowed the specimens to expand or contract freely during the thermal cycling.

In total 100 thermal cycles were applied to the test specimens. In total 3 specimens were tested in this experiment. Initiation of micro-damage (such as intralaminar cracks) during thermal cycling was monitored and quantified in 90-plies of every specimen by performing optical microscopy inspection after 5, 10, 20, 50 and 100 thermal loading cycles.

It was noted that only after 50 thermal loading cycles, a few intralaminar cracks similar to those in mechanical tensile tests initiate in the laminate plies at the edge. Delaminations were not found. Edge replication, described as option 3, was also used after 20 cycles, 50 cycles and 100 cycles, to open the cracks if they existed, as it is shown in Figure 9.

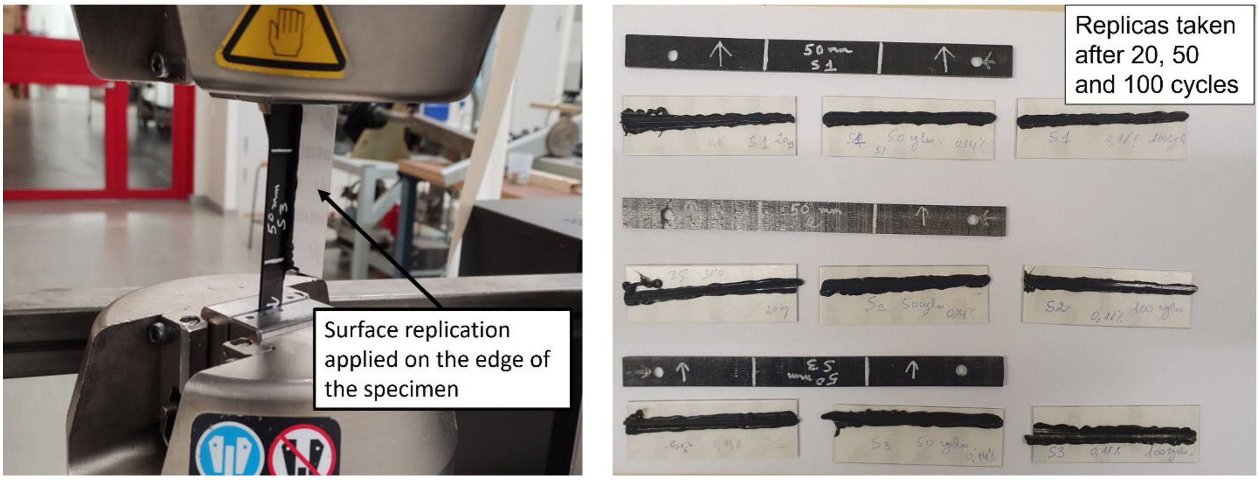

Figure 9 – Figure describing how replica was used.

Evolution of transverse crack initiation in laminate plies during cyclic thermal loading is presented in Table 2.

Table 2 – Evolution of transverse cracks in laminate layers during thermal cycling.

	Looking at the Replicas (length 50mm)			**Looking directly at the edge (length 50 mm)**
Specimens	**20 Cycles**	**50 Cycles**	**100 Cycles**	**100 Cycles**
Specimen 1	0	4	10	7
Specimen 2	0	0	2	2
Specimen 3	0	0	x	0

Edge replication was employed to facilitate crack visualization, as transverse cracks were not visible without this technique. A gauge length of approximately 50 mm was examined, resulting in the identification of only four cracks in specimen 1 after 50 cycles. Subsequently, a slight increase in the number of cracks was observed in both specimens 1 and 2 after 100 cycles. However, the edge replication quality for specimen 3 was not good, rendering it impractical for crack examination under the microscope.

5. Conclusions

Cross ply $[90/0]_{4s}$ composite laminates were manufactured using filament winding. The laminates were inspected, and specimens prepared for testing. Quasi-static, mechanical fatigue and thermal fatigue tests were performed. Only a few matrix cracks were observed at very high load and high number of cycles. Those cracks were initiated but not propagated along the width of the specimens. The test results showed that the material has a high damage onset level and therefore makes it an interesting candidate to make hydrogen composite tanks and pipes. The limited damage indicates that there are no obvious leak paths through the thickness that would compromise the functionality of the material in this application. The material durability under cryogenic operating temperatures looks promising.

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8. Copyright Statement

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