Effect of Thermal Cycling and Aging on Heated Fiber Metal Laminates and Glass-Fiber Epoxy Composites

Hagenbeek, Michiel; Müller, Bernhard; Sinke, Jos

DOI
10.1002/adem.201800084

Publication date
2018

Document Version
Accepted author manuscript

Published in
Advanced Engineering Materials

DOI: 10.1002/adem.201800084

Effect of thermal cycling and ageing on heated fibre metal laminates and glass-fibre epoxy composites**

By *Michiel Hagenbeek,** *Bernhard Müller,* and *Jos Sinke*

[*] *Dr. M. Hagenbeek, Dr. B. Müller, M.Sc. J. Sinke*

Aerospace Engineering, Delft University of Technology
Kluyverweg 1, 2629 HS Delft, The Netherlands
m.hagenbeek@tudelft.nl

[**] *This study is funded by the Dutch Technology Foundation NWO and Fokker Aerostructures.*

By integrating heater elements in Fibre Metal Laminates, such as GLARE, the material has a promising potential as de- or anti-icing system in aircraft structures. Application of this so called 'heated GLARE' material would however result in increased and (up to ten fold) more frequent temperature loading compared to the regular flight conditions.
To investigate the long-term effects thermal cycling tests were performed up to 36000 cycles in three different temperature ranges. Both heated GLARE and FM906 glass-fibre epoxy samples have been thermal cycled using an in-house developed thermal cycling setup. Heating was performed in two different ways, from the outside using Peltier elements and from the inside using integrated heater elements. In addition, continuous heating tests were performed to further examine the effect of ageing and internal stress relaxation.
FM906 glass-fibre epoxy samples cycled by external heating showed an increase in interlaminar shear strength (ILSS) as a result of physical ageing, whereas internal heating showed a decrease. Most GLARE samples cycled by internal heating showed an increase in ILSS. The continuous heating tests on heated GLARE confirm that ageing and internal stress relief have counteracting effect on the ILSS. The temperature level determines which effect is most dominant.

1. Introduction

Fibre Metal Laminates, such as GLARE [1], have been successfully introduced in the A380 fuselage and leading edges. The material offers improved damage tolerance and a significantly reduced crack growth compared to conventional aluminium alloys.[2,3] By integrating heater elements the material has a promising potential as de- or anti-icing system in aircraft structures. Such a heated GLARE variant would however be exposed to higher and up to ten fold more frequent temperature loading of the material compared to the regular flight conditions. For deicing conditions the temperature range typically is -25 to 80°C [4,5], whereas flight conditions range between -55 to 70°C. [6] The durability of the material under these higher and intensified thermal loadings needs to be investigated to meet the high quality standards from the aircraft industry.

The long-term effects on the material under these thermal loading conditions can be simulated by means of thermal cycling tests. Park found 9 to 18% reduction in interlaminar shear strength (ILSS) for unconditioned GLARE 5-7/6-0.4 specimens after 1500 thermal cycles between 25 to 125°C with each 15 min duration time. [7] A smaller 8 to 13% decrease in ILSS for glass-fibre epoxy $[0]_{40}$ was found under the same conditions. Park explains this by internal stresses in the GLARE specimens that the UD glass-fibre epoxy specimens do not have. Whereas GLARE 2/1-1.6 tested by Da Costa did not show tensile or interlaminar shear strength reduction after thermal cycling between -50 and +80°C for up to 2000 cycles. [8]

A difference between Park and Da Costa is the aluminium thickness (Park used 0.4 mm and Da Costa 1.6 mm). The latter is expected to give a more gradual heating. Another difference is the fact that Da Costa performed the thermal cycling up to 80°C, around the glass-transition temperature (Tg), and Park up to 125°C (well beyond Tg). The lower temperatures and larger temperature range of Da Costa (-50 to 80°C) compared to Park (25 to 125°C) seem to play a less important role here than the maximum temperature, since no strength reduction was found by Da Costa. It should be noted that in both the investigation of Park and of Da Costa standard GLARE based on FM94 glass-fibre epoxy (Tg=103°C) is used. In the current research FM906 glass-fibre epoxy (Tg=135°C) is used.

Residual thermal stresses exist in GLARE due to the different thermal expansion coefficients of the GLARE constituents. Analytical calculations confirm that these stresses do not rise significantly when the temperature of GLARE specimens is gradually decreased from room temperature to lower temperatures, when taking into account the change in thermal expansion coefficient and stiffness of the glass-fibre epoxy layers with temperature. On the other hand, at elevated temperatures there is a significant drop in the epoxy matrix stiffness around Tg (see **Figure 1**). Thus, the thermal residual stresses change significantly more when cycling from room temperature beyond the Tg of the polymer matrix. This indicates that the material Tg plays an important role on the effect of thermal cycling. Besides the large change in internal stress, cycling beyond Tg might also cause molecular change in the epoxy or affect the interfaces between the different constituents. To investigate the effect of local heating on the fibre metal laminate numerical modeling can be used as shown by Jiang. [9,10]

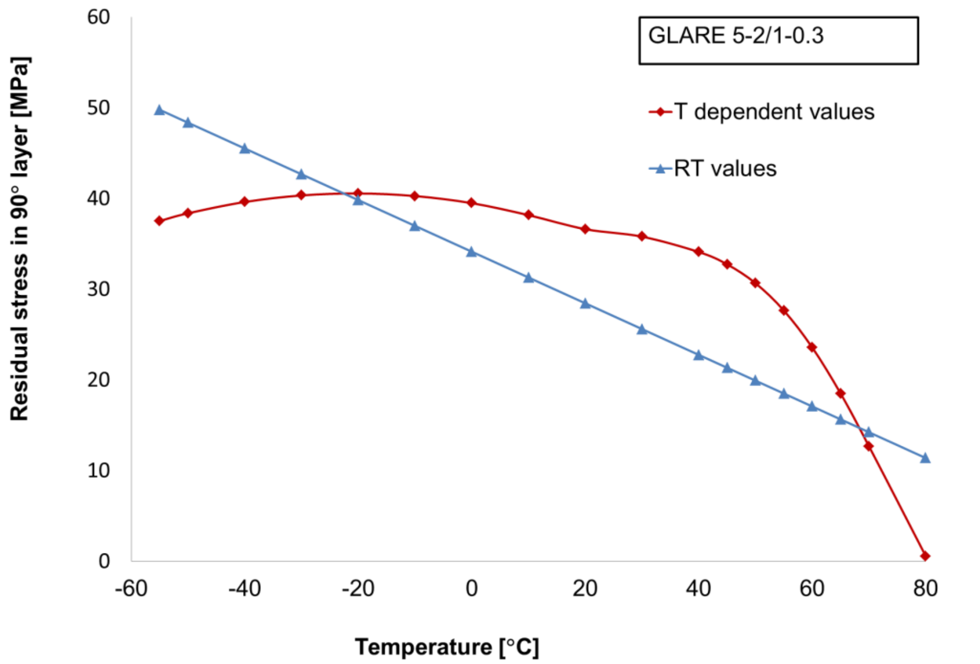


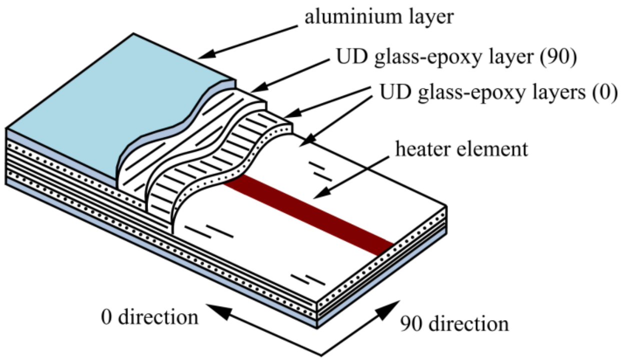
Figure 1. Residual stress calculation in the 90° FM94 glass-fibre epoxy layer of a GLARE 5-2/1-0.3 based on either constant (room temperature) or temperature dependent properties.

In this paper an in-house developed thermal cycling setup has been used to perform tests up to 36000 cycles to investigate the effect of thermal cycling further, and beyond 2000 cycles. This number is based on the assumption that deicing is applied once during both ascend and descend in 20% of the 90.000 flights, which is the design service life for a regional liner. It should be noted however that in reality the heating for de-icing is not applied continuously to the structure but via short heat cycles every two or three minutes. Thus the total number of cycles could easily be tenfold, when systems are turned on half an hour during icing conditions. As a first step in extended thermal cycling the current research was limited to 36000 cycles (36kC). Both heated GLARE samples and FM906 glass-fibre epoxy samples (with and without heater element) have been thermal cycled. The heating was performed in two ways: from outside via the Peltier elements and from inside using integrated heater elements. In addition, continuous heating tests have been performed in two different ways: through oven heating for FM906 glass-fibre epoxy samples without heater element and by means of internal (resistance) heating for heated GLARE samples (with an integrated heater element).

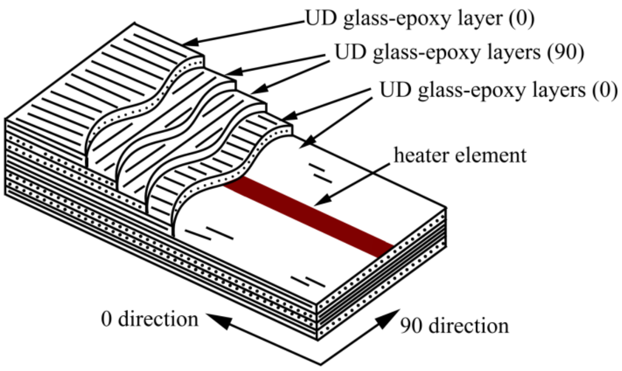
2. Heated GLARE

2.1. The heated GLARE lay-up

A heated GLARE variant was developed to integrate a heating function in the certified (structural) aircraft material GLARE. [11] Heated GLARE is expected to be applied as a de-/anti-icing device in leading edges of aircraft. GLARE is a FML which consists of alternating 2024-T3 aluminium and unidirectional (UD) FM94 glass-fibre reinforced epoxy (GFRP) layers. [11] The heating function is provided by embedding copper heater elements between the UD GFRP layers. Similarly, a heated full composite cross-ply laminate, i.e. with the same UD GFRP layers but without aluminium layers, can be build up. **Figure 2** schematically depicts such a heated GLARE layup and heated full composite cross-ply layup. The two different layups were subjected to thermal cycling in the current research to investigate both the material durability and the influence of the aluminium layers.



(a)



(b)

Figure 2. Schematic lay-up of a heated GLARE laminate (a) and a heated full composite cross-ply laminate (b).

Two glass-fibre epoxy prepregs, FM94 or FM906, are used for GLARE. FM94 is the standard epoxy which is used for conventional GLARE. FM94 is cured at a temperature of 120°C for one hour in the autoclave at 6 bar. In case FM906 epoxy is used in combination with aluminium 7475-T761, the FML is referred to as high static strength (HSS) GLARE which has improved strength and service temperature over the standard GLARE. [12] The higher allowable service temperature results from the higher glass transition (Tg = 135°C) and curing (180°C) temperatures. FM906 glass-fibre prepreg epoxy is used in heated GLARE as temperatures up to 80°C are expected and the stiffness of the FM94 significantly decreases at temperatures beyond 70°C. [13] Different types of aluminium can in principle be used, however all heated GLARE laminates in the current research are based on aluminium 2024-T3.

2.2. Loading conditions of the leading edge

A heated leading edge is subjected to mechanical and thermal loading conditions. Both the mechanical and the thermal loading have a cyclic nature and can lead amongst others to fatigue damage. The event of an impact, for example due to bird strike, can lead to impact damage on the leading edge. During the lifetime of the structure moisture ingress can take place along the edges and bore holes. The amount of moisture ingress is expected to be limited to 0.2% as shown by Beumler. [14] In the current study only the effect of cyclic thermal loading on (heated) GLARE and glass-fibre epoxy composite is investigated.

Thermal stresses are present during operation due to temperature changes during ascents and descents (once per flight) [6] and in case of heated leading edges which are used as local anti- and de-icing devices (several times per flight). [4, 5] Other reasons for thermal (residual) stresses are the inhomogeneous temperature distributions which especially occur when the embedded heater elements are switched on and off [15, 16], and due to the different thermal expansion coefficients of the used materials (aluminium, FM906 epoxy, glass-fibres and copper). The elevated temperatures during icing conditions are expected to cause physical ageing of the epoxy too. [17]

3. Experimental procedures

3.1. Material manufacturing

All heated GLARE and heated full composite cross-ply laminates in the current research were manufactured using FM906 glass-fibre epoxy prepreg. The manufacturing procedure for heated GLARE consists of three main steps. The first step is cutting and laminating of the aluminium, the copper heater element and the prepreg layers at room temperature in a clean room.

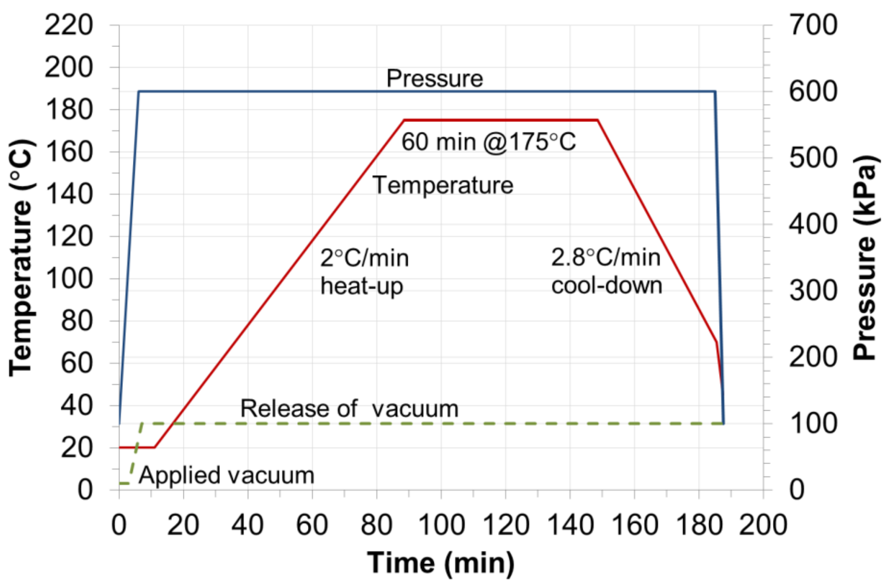


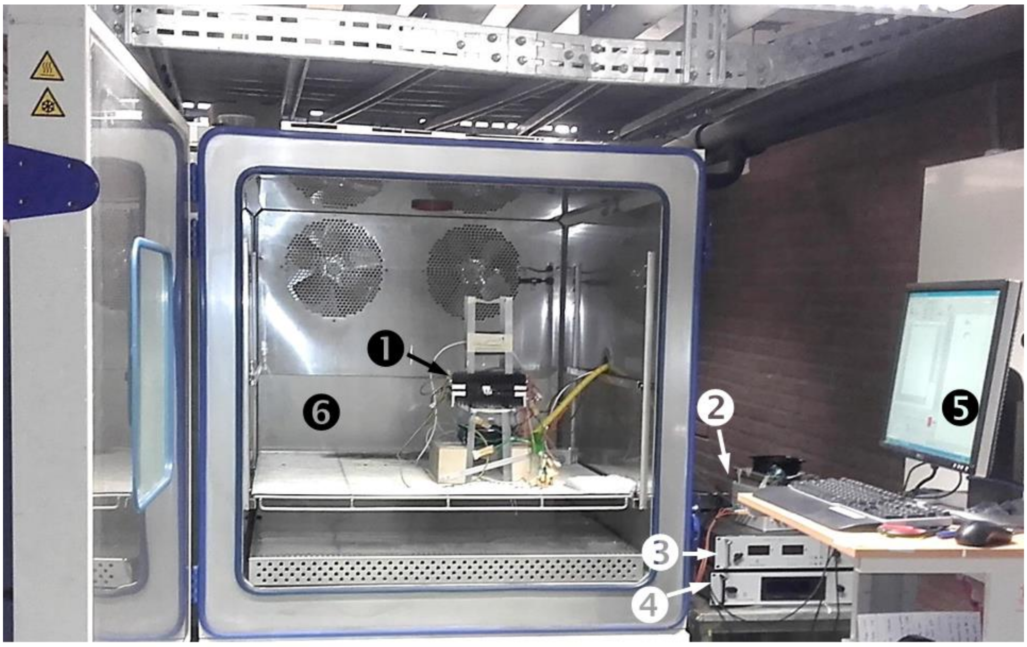
Figure 3. Autoclave cycle for 180°C curing FM906 glass-fibre epoxy as used in heated GLARE.

The next step is applying vacuum to the panels using a vacuum bag. In the final step the panels are cured in the autoclave at a temperature of 180°C and a pressure of 6 bar for one hour. [6] **Figure 3** shows the vacuum, pressure and temperature set points which are used for manufacturing of HSS GLARE. For the heated full composite cross-ply laminates the same manufacturing procedure as for heated GLARE is followed.

3.2. Thermal cycling setup

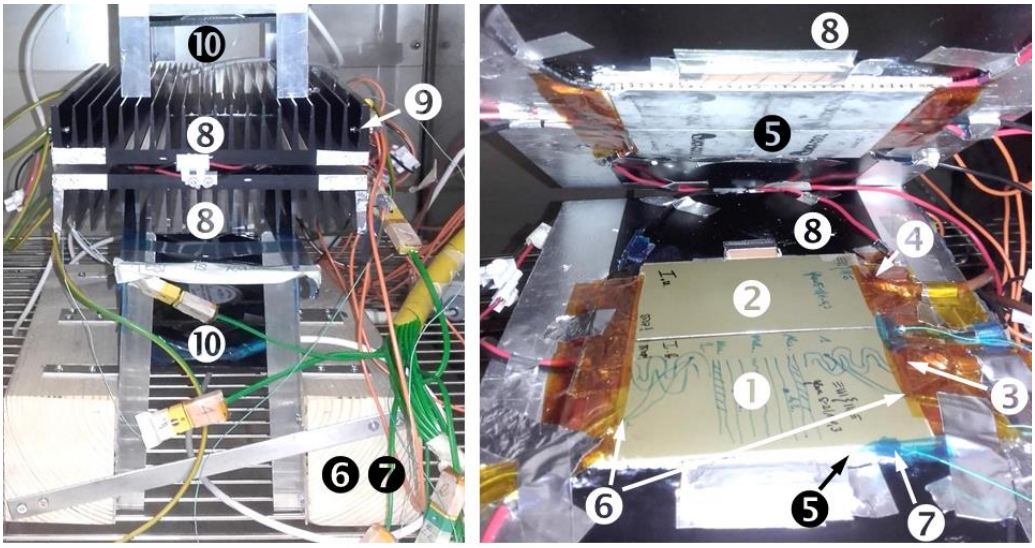
The thermal cycling machine used for the thermal fatigue tests was developed within the heated GLARE project. A detailed description of the thermal cycling machine and results of thermal fatigue tests up to 12.000 cycles, with and without embedded heater elements, have already been reported. [18,19] An overview of the thermal cycling setup is shown in **Figure 4**. The design of the innovative thermal cycling machine was motivated by the requirement to cycle both conventional GLARE and heated GLARE with the same setup. Thus, two different means of thermal cycling (heating) were required. For heated GLARE the machine had to allow for external cooling and internal heating. For conventional GLARE the machine had to allow for external cooling and external heating. The internal heating was achieved through resistance heating by applying direct current (DC) to the embedded mesh. This corresponds with its original use as anti-/de-icing device. [11]

The external heating and external cooling was realised using Peltier elements on both sides of the sample. Peltier elements cool on one side and heat on the opposite side when DC is applied. [20] When the direction of the current is changed, the sides which heat and cool swap. Thus, the samples get thermal cycled by changing the DC direction when predefined sample temperatures are reached. The temperatures which are used to control the switching of the DC direction are measured in the samples using embedded thermocouples. The switching of the DC current is done by a relais which is controlled by the computer.



❶	Thermal cycling machine	❹	Power supply 2
❷	Box with relay	❺	Computer
❸	Power supply 1	❻	Climate chamber

(a)



❶	Sample 1	❻	Embedded thermocouples
❷	Sample 2	❼	Thermocouples at the edges
❸	Electrical input	❽	Heat sinks
❹	Electrical output	❾	Thermal switches
❺	Peltier elements	❿	Ventilators

(b)

Figure 4. Photos of the thermal cycling machine in the climate chamber (a) overview and (b) detail.

By using Peltier elements the sample is not exposed to cooling liquids. Thus, related effects of the cooling liquid on material composition or moisture ingress are avoided. Another advantage of the setup is that the temperature range and temperature profile are adapted by the electrical heating and cooling power, which can be computer controlled.

The minimum temperature can be reduced further by placing the thermal cycling setup into a climate chamber with low temperature. By switching the external cooling on (constant cooling power or intermittent cooling power) and frequently switching the internal heating on and off, anti- and de-icing conditions of heated leading edges can be simulated.

3.3. Thermal cycling samples

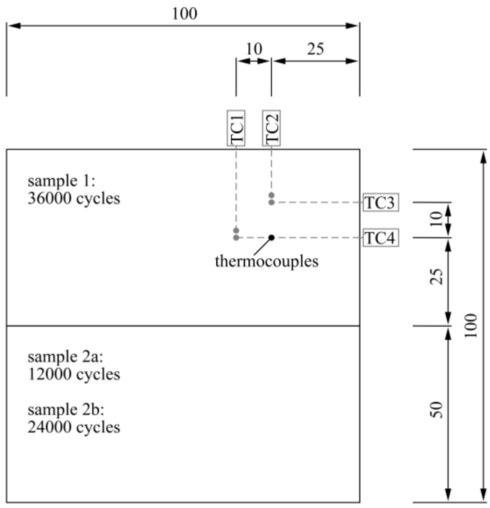
Sixteen samples were thermal cycled in this study. The layups of all these samples are depicted in **Table 1**. The specimens with the number 0 in their abbreviation were not thermal cycled and used as a reference, all other samples were thermal cycled. The samples with the abbreviations GE0 to GE3 (glass-fibre epoxy) consist of 8 glass-fibre epoxy layers. The letter H before the abbreviation GE indicates that heater elements according to **Figure 5** were embedded in the samples HGE0 to HGE4. The samples with the abbreviations HGL0 to HGL6 indicate a heated GLARE 5-2/1-0.3 layup, i.e. they consisted of two 0.3 mm thick aluminium layers and four glass-fibre epoxy layers with fibre orientations of [0/90/90/0]. **Figure 5** shows the three different sample configurations that were thermal cycled in this study. The samples can be distinguished into samples without and with embedded heater elements. The sample configuration shown in (a) consists of two-part samples without heater elements that were thermal cycled using external cooling and external heating. The sample configuration as shown in (b) has embedded heater elements and are internal heated through resistance heating. Both the configurations as shown in (a) and (b) have been used for thermal cycling of FM906 glass-fibre epoxy composites.

Table 1. Samples: Nomenclature (nom), materials, thermal loading conditions, and number of cycles (noc).

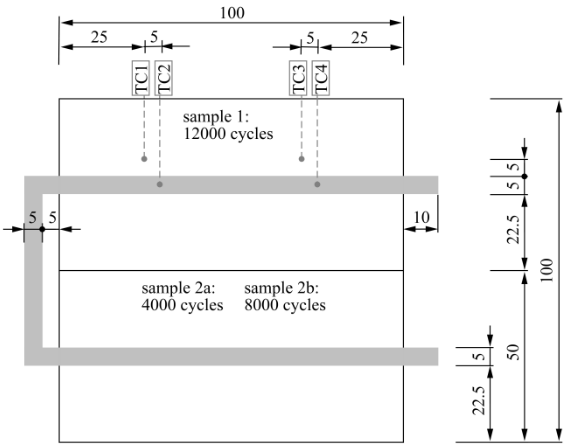
nom	Material & layup	T_{min} [°C]	T_{max} [°C]	noc [-]
GE0	FM906 $(0/90)_{2S}$	na	na	0
GE1	FM906 $(0/90)_{2S}$	0	120	12000
GE2	FM906 $(0/90)_{2S}$	0	120	24000
GE3	FM906 $(0/90)_{2S}$	0	120	36000
HGE0	FM906 $(90/0/0/90)_{2S}$	na	na	0
HGE1	FM906 $(90/0/0/90)_{2S}$	-25	50	4000
HGE2	FM906 $(90/0/0/90)_{2S}$	-25	50	8000
HGE3	FM906 $(90/0/0/90)_{2S}$	-25	50	12000
HGE4	FM906 $(90/0/0/90)_{2S}$	-25	72.5	12000
HGL0	heated GLARE 5-2/1-0.3	na	na	0
HGL1	heated GLARE 5-2/1-0.3	-25	50	4000
HGL2	heated GLARE 5-2/1-0.3	-25	50	8000
HGL3	heated GLARE 5-2/1-0.3	-25	50	12000
HGL4	heated GLARE 5-2/1-0.3	-25	72.5	4000
HGL5	heated GLARE 5-2/1-0.3	-25	72.5	8000
HGL6	heated GLARE 5-2/1-0.3	-25	72.5	12000

By cutting a sample with an embedded heater element along the centre line four samples with identical size are obtained as depicted in **Figure 5 (c)**. This sample configuration has been used for thermal cycling of heated GLARE 5-2/1-0.3. Besides cycling multiple samples at the same time, this configuration allows for microscopic inspection and comparison of the interfaces before and after testing.

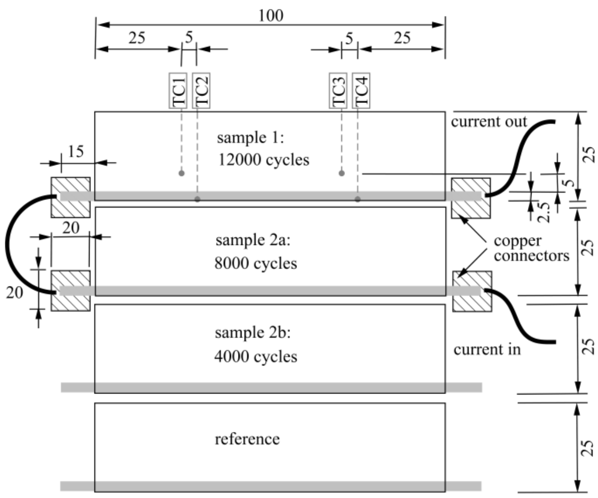
Two-part samples were used as they allow for an economic use of the thermal cycling machine. In the first stage the samples 1 and 2a were put in the thermal cycling machine and cycled. After *m* thermal cycles the sample 2a was removed and the sample 2b was inserted. Then the thermal cycling was continued for another *n* thermal cycles. Thus, sample 1 was exposed to a total number of $m+n$ thermal cycles while the samples 2a and 2b were exposed to *m* and *n* cycles respectively.



(a)



(b)



(c)

Figure 5. Dimensions and arrangement of the samples 1, 2a and 2b for materials (a) without heater mesh, and (b, c) with heater mesh.

Consequently, the overall thermal cycling time was reduced by one half. With the four-part samples the intermediate levels of cycling could be increased even further with a reduction of the overall thermal cycling time to a quarter compared to sequential testing. In the current research this option was not used.

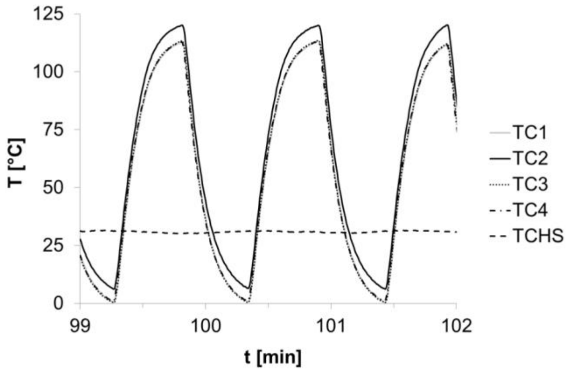
The heater element of sample configuration as shown in **Figure 5 (b)** was positioned in the centre plane of the sample to enable a symmetric thermal loading of the samples. Straight copper stripes with a width of 5 mm and a thickness of 0.12 mm were used as heater elements. Straight elements are the simplest possible heater elements and they eliminate geometry effects on the stress distributions. Furthermore, straight heater elements enable cutting of identical specimens for the mechanical tests. The heater element as shown in sample configuration (c) has an approximate width of 2.5 mm. Four thermocouples were embedded in the samples to control and monitor the thermal loading of the samples. In sample 1 two thermocouples were embedded in the centre plane of the sample (TC1, TC3) and two between the second and third glass-fibre epoxy layers (TC2, TC4). The in-plane positions of the thermocouples are depicted in **Figure 5**.

3.4. Thermal cyclic fatigue testing

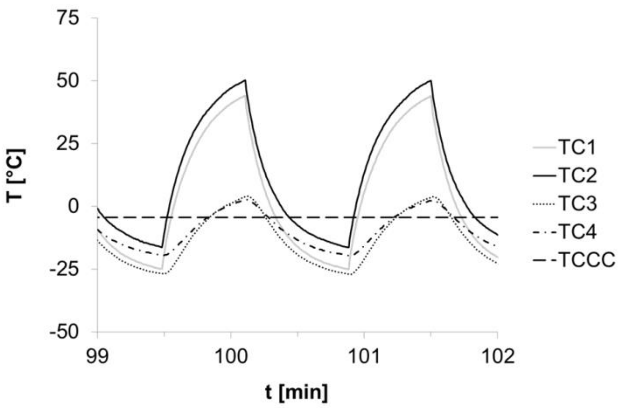
The GLARE and glass-fibre epoxy samples were cycled in three different temperature ranges. The $[0/90]_{2s}$ laminates with a thickness of 1 mm (GE1-GE3) were cycled using the Peltier elements for both external heating and external cooling. The temperature range was 0°C to 120°C. The latter corresponds with the maximum service temperature of the FM906 epoxy. Physical ageing of the epoxy is known to be present at those elevated temperatures from previously conducted tests. [18]

Previous tests however ran up to 12000 cycles, which was chosen here as the starting point. The current research proceeds with an intermediate level of 24000 cycles and a total number of 36000 cycles as end point. This high number of cycles correspond with the expected number of cycles during the service life of aircraft as mentioned earlier. **Figure 6 (a)** shows the measured temperatures at the thermocouple positions indicated in **Figure 6 (a)** during a typical 0°C to 120°C thermal cycle. The relatively small thickness of 1 mm leads to small temperature gradients across the thickness. As a result, physical ageing is expected to occur homogeneously across the sample thickness.

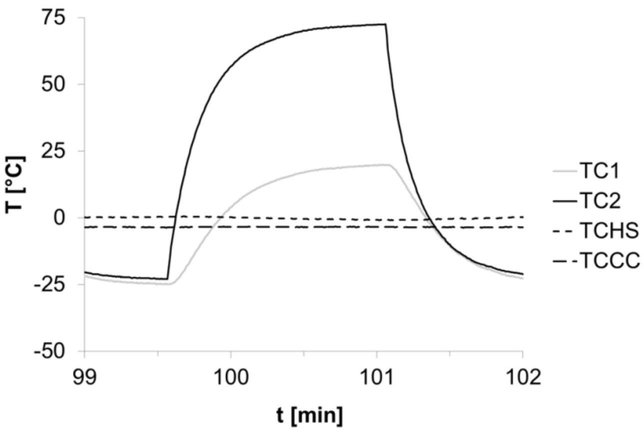
The samples with integrated heater elements (HGE1-HGE3, HGL1-HGL3) were cycled between -25°C to 50°C to avoid distinct physical ageing of the samples due to elevated temperatures. Thus, the focus was specifically on the possible effects on the material properties induced by constantly changing thermal residual stresses during the thermal cycling. Thermal residual stresses result from the manufacturing conditions at 180°C and increase with decreasing temperatures. As the heating was performed using the internal mesh and the cooling was performed (externally) using the Peltier elements, the thermal stresses decrease during the heating phase in the vicinity of the heater elements. [16] The number of cycles were chosen according to previous studies. [18] Figure 6 (b) shows the typical thermal cycle of the heated FM906 glass-fibre epoxy samples (HGE1-HGE3) in the temperature range of -25°C to 50°C. **Figure 7 (a)** shows the typical heated GLARE 5-2/1-0.3 thermal cycle in the same temperature range of -25°C to 50°C (HGL1-HGL3). The temperature of the heat sink (TCHS) and the climate chamber (TCCC), are depicted in **Figure 6** and **7** as well. The temperature of the heat sink is measured to control the heat coming from the back side of the Peltier elements, see **Figure 4**. The samples HGE4, HGL4, HGL5, and HGL6 were thermal cycled in the range of -25°C to 72.5°C with internal heating using the heater mesh and external cooling using the Peltier elements. **Figure 6 (c)** and **Figure 7 (b)** shows the typical thermal cycles of the heated FM906 glass-fibre epoxy sample (HGE4) and the heated GLARE 5-2/1-0.3 samples (HGL4-HGL6) in this case.



(a)

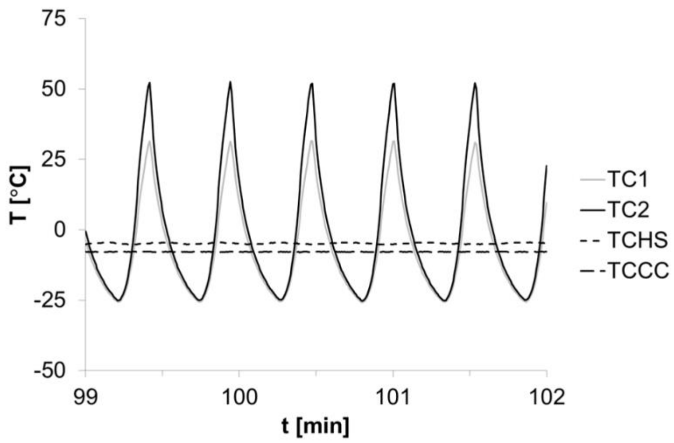


(b)

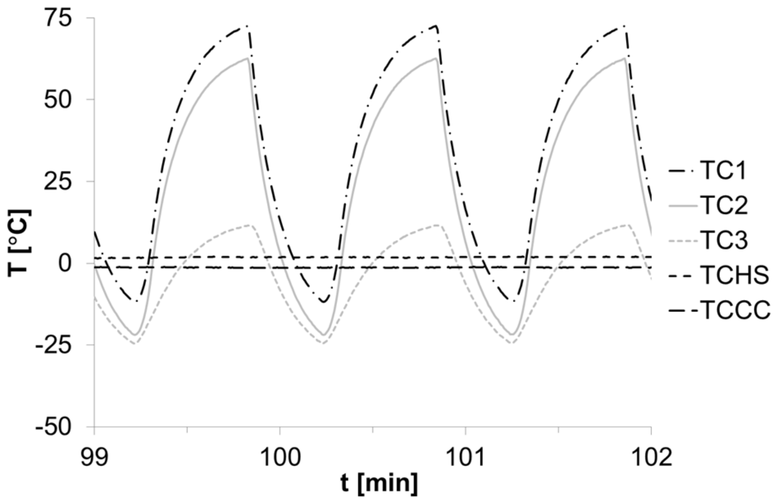


(c)

Figure 6. Typical (heated) FM906 glass-fibre epoxy thermal cycles of (a) GE1 to GE3, (b) HGE1 to HGE3 and (c) HGE4 (cf. Table 1) measured at thermocouple (TC) positions, see Fig. 5. Temperature of heat sink (TCHS) and climate chamber (TCCC) are shown as well.



(a)



(b)

Figure 7. Typical heated GLARE 5-2/1-0.3 thermal cycles of (a) HGL1 to HGL3, (b) HGE4 to HGL6 (cf. Table 1) measured at the thermocouple (TC) positions depicted in Figure 5. Temperature of the used heat sink (TCHS) and climate chamber (TCCC) are depicted as well.

All internal heated samples were cycled for 4000, 8000, and 12000 cycles. **Table 2** shows the total exposure time (after 12000 cycles) of the samples in hours for each ten degree temperature step. The (total) time at elevated temperature is expected to cause physical ageing of the samples. This effect has been observed for FM906 glass-fibre epoxy cross-ply composites that were thermal cycled between 0°C and 120°C for 12000 cycles. [19]

Table 2. Total time in hours of the specimens which were exposed to 12000 thermal cycles (cf. Figures 6 and 7). The cycle time and the min. and max. temperature are given as well.

Temp. range [°C]	GE1 [h]	HGE3 [h]	HGE4 [h]	HGL3 [h]	HGL6 [h]
-30 to -20	0.0	60.0	105.0	25.0	21.7
-20 to -10	0.0	45.0	81.7	23.3	13.3
-10 to 0	0.0	23.3	31.7	15.0	10.0
0 to 10	25.0	13.3	23.3	10.0	18.3
10 to 20	26.7	23.3	20.0	8,3	15.0
20 to 30	18.3	25.0	1.7	6.7	15.0
30 to 40	11.7	35.0	21.7	6.7	13.3
40 to 50	11.7	55.0	23.3	10.0	16.7
50 to 60	11.7	0.0	36.7	0.0	23.3
60 to 70	10.0	0.0	90.0	0.0	36.7
70 to 80	8.3	0.0	113.3	0.0	21.7
80 to 90	11.7	0.0	0.0	0.0	0.0
90 to 100	11.7	0.0	0.0	0.0	0.0
100 to 110	18.3	0.0	0.0	0.0	0.0
110 to 120	50.0	0.0	0.0	0.0	0.0
Σ	215.0	280.0	548.3	105.0	205.0
Min temp [°C]	0	-25	-25	-25	-25
Max temp [°C]	120	50	72.5	50	72.5
Cycle time [*s*]	64.5	84.0	164.5	31.5	61.5

3.5. Continuous heating samples and test setup

In addition to the thermal cycling tests two different continuous heating tests were performed to further examine the effect of elevated temperature on the used material. In the first test FM906 glass-fibre epoxy samples, without integrated heater element, were put into an oven (Votch 0951). The samples were air-tight packed in kapton tape to prevent possible material oxidation to the air. The used temperature level was 120°C and the set times ranged from 24, 48 to 96 hours.

In the second test heated GLARE 5-2/1-0.3 samples were internal heated using a power supply (Delta electronics SM1540). The current was manually controlled to keep the sample at the desired temperature level, 50, 80 and 120°C, and the set times; 24, 48, 96, and 192 hours.

Table 3. Samples: Nomenclature (nom), materials, thermal loading conditions, ageing times.

nom	Material & layup	T_{level} [°C]	Ageing times [hours]
AGE0	FM906 $(0/90)_{2S}$	na	0
AGE1	FM906 $(0/90)_{2S}$	120	24
AGE2	FM906 $(0/90)_{2S}$	120	48
AGE3	FM906 $(0/90)_{2S}$	120	96
AHGL0	heated GLARE 5-2/1-0.3	na	0
AHGL1	heated GLARE 5-2/1-0.3	50	24
AHGL2	heated GLARE 5-2/1-0.3	80	24
AHGL3	heated GLARE 5-2/1-0.3	120	24
AHGL4	heated GLARE 5-2/1-0.3	50	48
AHGL5	heated GLARE 5-2/1-0.3	80	48
AHGL6	heated GLARE 5-2/1-0.3	120	48
AHGL7	heated GLARE 5-2/1-0.3	50	96
AHGL8	heated GLARE 5-2/1-0.3	80	96
AHGL9	heated GLARE 5-2/1-0.3	120	96
AHGL10	heated GLARE 5-2/1-0.3	50	192
AHGL11	heated GLARE 5-2/1-0.3	80	192
AHGL12	heated GLARE 5-2/1-0.3	120	192

The test matrix for the two different heating tests is shown in **Table 3**. The used abbreviation in the sample nomenclature is similar to those used for the thermal cycled samples. The letter A before the abbreviation GE and HGL indicates the ageing of the samples in this case. The samples with number 0 are non-aged and used as a reference. The used material is identical to the material used in the thermal cycling tests.

3.6. Interlaminar shear strength testing

Interlaminar shear strength (ILSS) tests were performed in threefold on all samples listed in **Table 1** according to the ASTM standard. [21] The specimens have a width of 4 mm and a length of 20 mm. All ILSS samples were taken from the position of the heater element if not explicitly stated otherwise. The ILSS tests were conducted on a 25 kN test machine with a test speed of 1 mm/min. Correct shear failure modes were obtained in all tests.

Figure 8 shows the ILSS test results for the FM906 glass-fibre epoxy specimens GE1 to GE3 thermal cycled between 0 and 120°C by external heating in comparison with the reference (non-cycled) GE0 specimens.

The ILSS test results for the aged FM906 glass-fibre epoxy specimens AGE1 to AGE3 are given in **Figure 9**. It shows an increasing interlaminar shear strength for longer exposure to elevated temperature. In **Figure 10** the ILSS results for the heated FM906 glass-fibre epoxy specimens HGE1 to HGE3 thermal cycled between -25 and 50°C by internal heating and external cooling in comparison with the reference (non-cycled) HGE0 specimens are given.

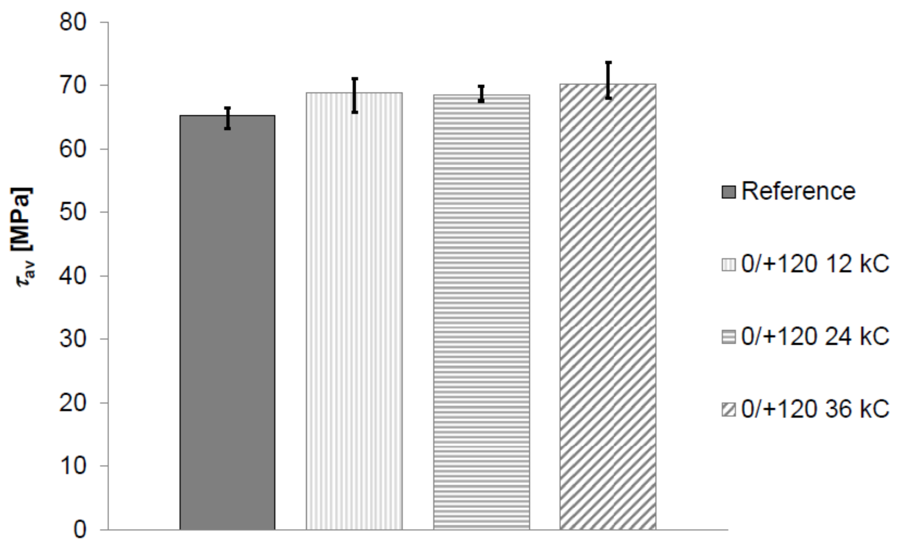


Figure 8. FM906 glass-fibre epoxy specimens GE1 to GE3 thermal cycled between 0 and 120 °C by external heating compared to the reference (non-cycled) GE0 specimens (cf. Table 1)

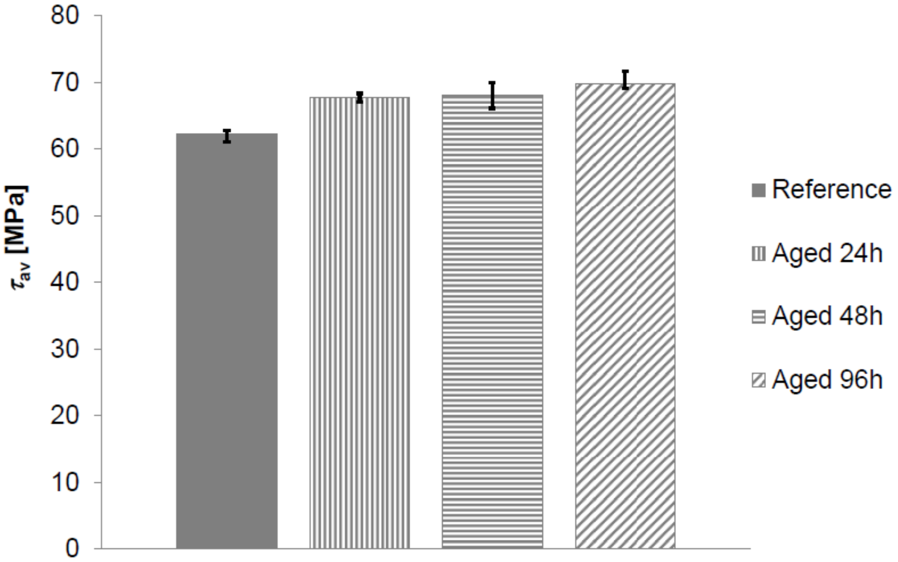


Figure 9. FM906 glass-fibre epoxy specimens AGE1 to AGE3 aged in the oven at 120°C compared to the reference (non-aged) AGE0 specimens (cf. Table 3).

In **Figure 11** the ILSS results of the heated FM906 glass-fibre epoxy specimens HGE4, thermal cycled between -25 and 72.5°C, are given and compared to the specimens HGE1 to HGE3, thermal cycled between -25 and 50°C, and the reference (non-cycled) HGE0 specimens.

The ILSS test results for heated GLARE 5-2/1-0.3 specimens HGL1 to HGL3, thermal cycled between -25 and 50°C, and specimens HGL4 to HGL6, thermal cycled between -25 and 72.5 ∘C, in comparison with the reference (non-cycled) HGL0 specimens are given in **Figure 12**.

The ILSS test results for the continuously heated GLARE 5-2/1-0.3 specimens AHGL1 to AHGL12 are given in **Figure 13**. These specimens were internal heated to a steady state temperature of 50, 80 and 120°C for 24, 48, 96 and 192 hours. Where both the 80°C and 120°C show a slight decrease after the initial drop at 24 hours, the 50°C specimens show a much more pronounced decrease for 96 and 192 hours of internal heating.

The edges of the heated GLARE samples HGL3 and HGL6 were polished before cycling for 12kC in the temperature range of -25°C to 50°C and -25°C to 72.5°C respectively.

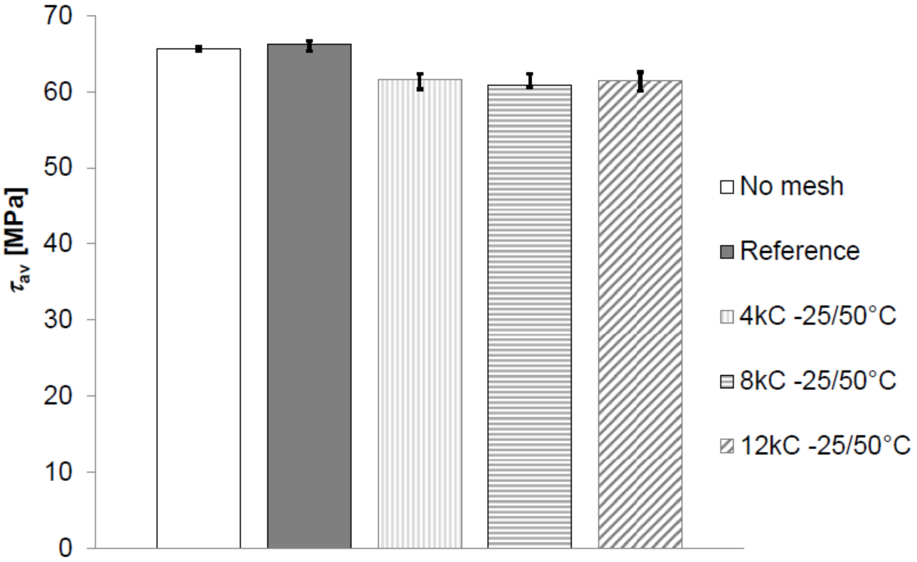


Figure 10. Heated FM906 glass-fibre epoxy specimens HGE1 to HGE3 thermal cycled between -25 and 50°C by internal heating compared to the reference (non-cycled) HGE0 specimens (cf. Table 1).

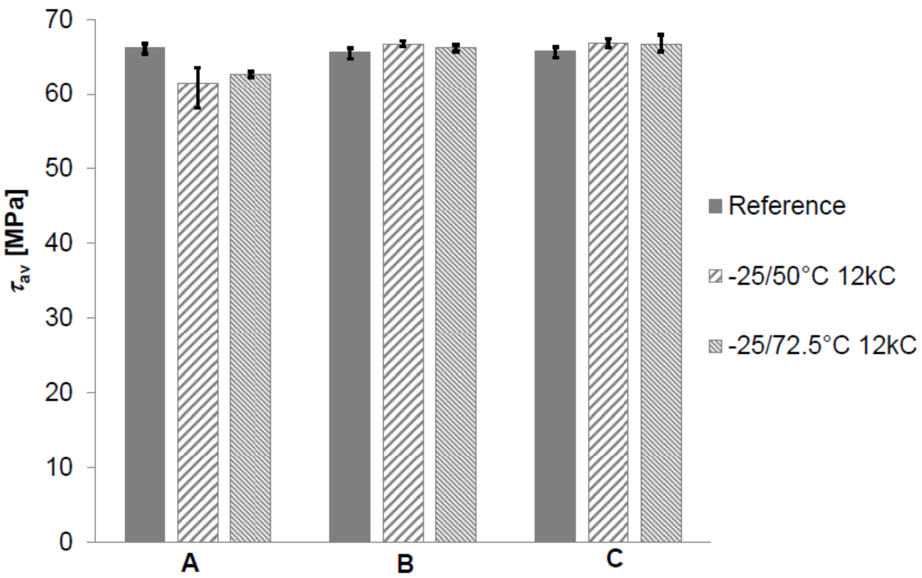


Figure 11. Heated FM906 glass-fibre epoxy specimens thermal cycled between -25 and 50°C (HGE3) and between -25 and 72.5°C (HGE4) compared to the reference (non-cycled) HGE0 specimens (cf. Table 1). ILSS specimens taken from the heater element position (A), right beside the heater element (B), and 5 mm from the heater element edge (C).

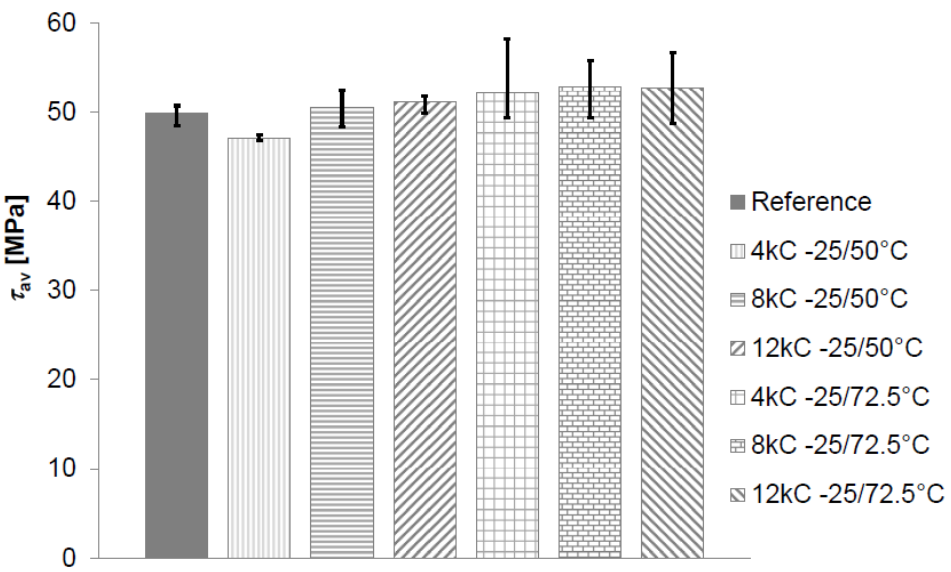


Figure 12. Heated GLARE 5-2/1-0.3 specimens HGL1 to HGL3 thermal cycled between -25 and 50°C and specimens HGL4 to HGL6 thermal cycled between -25 and 72.5°C compared to the reference (non-cycled) HGL0 specimens (cf. Table 1).

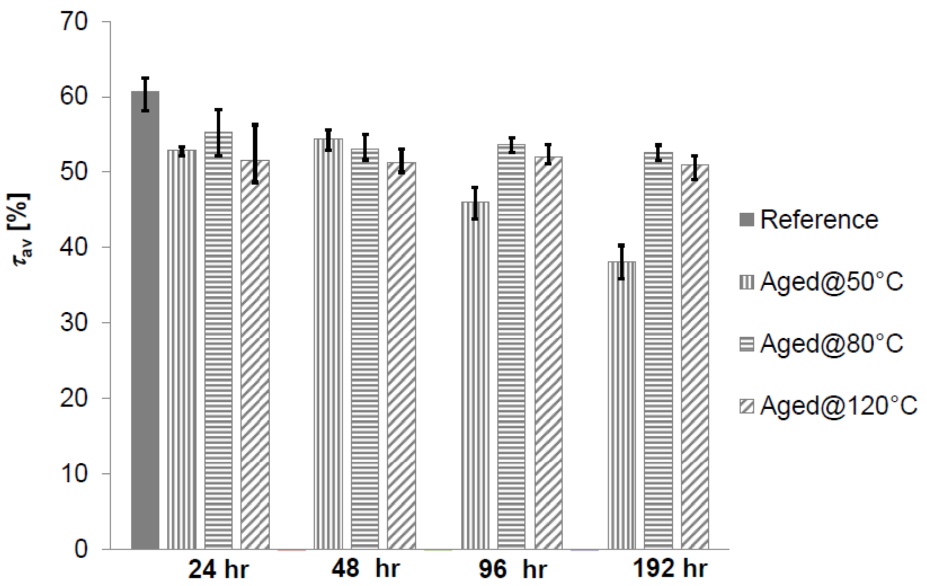
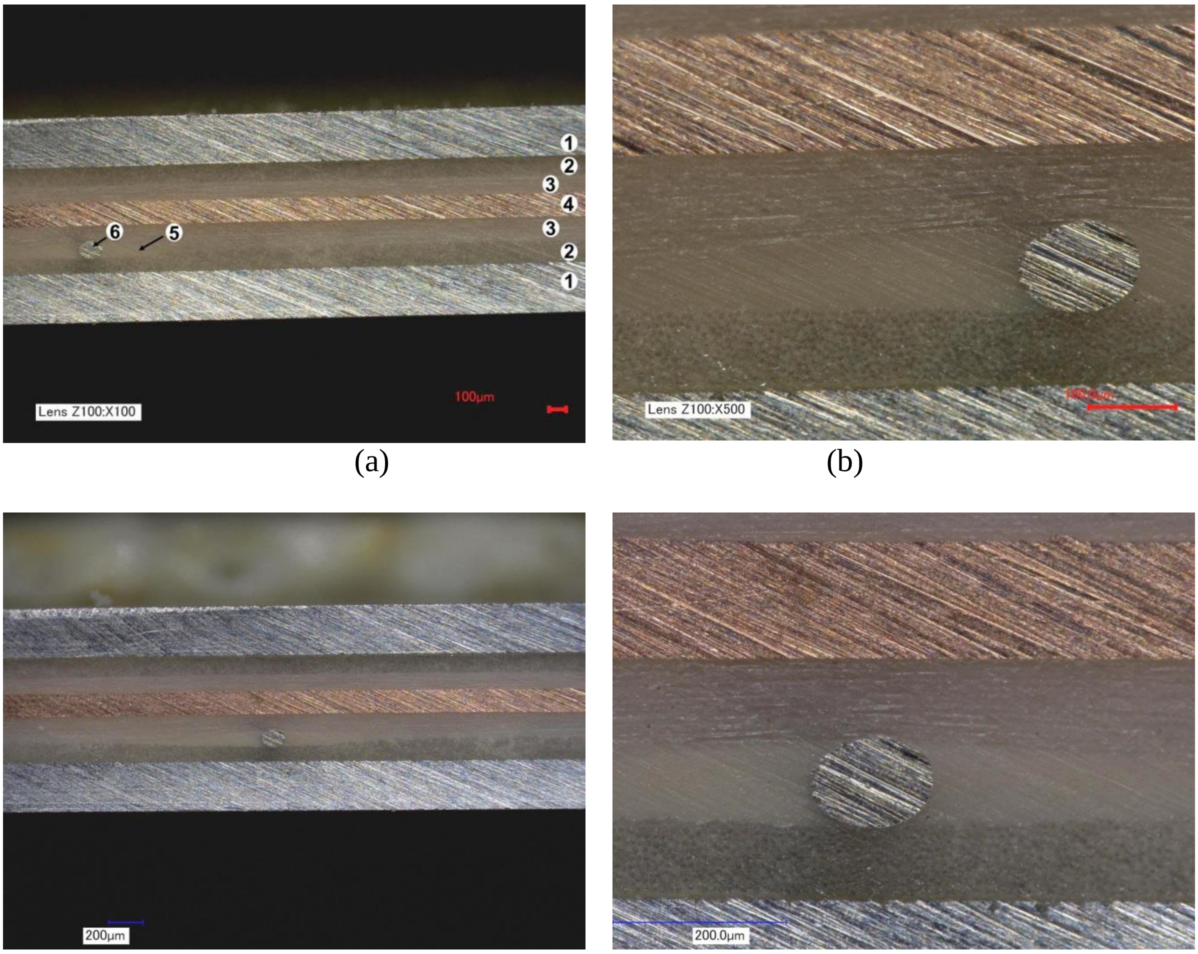


Figure 13. Heated GLARE 5-2/1-0.3 specimens AHGL1 to AHGL12 internal heated to 50, 80 and 120°C for 24, 48, 96 and 192 hours comparison to the reference (non-aged) AHGL0 specimens (cf. Table 3).

Microscopic imaging was then used to examine all materials and their interfaces before and after thermal cycling. **Figure 14** shows the optical microscope images with magnifications of 100x and 500x for sample HGL6 before (a,b) and after thermal cycling (c,d). Both sample HGL3 and HGL6 did not show any cracks, voids or other visible changes due to thermal cycling for 12kC.



(a) (b)

(c) (d)

Figure 14. Optical microscope images of heated GLARE sample HGL6, magnification of 100x and 500x, before (a, b) and after thermal cycling for 12kC from -25°C to 72.5°C (c, d). In (a) the aluminium (1), glass-fibre epoxy in 90° (2) and 0° (3), and copper heater element (4) are indicated, as well the resin rich area (5) created by the thermocouple (6).

4. Results and discussion

An increase in interlaminar shear strength up to 7.5% is found in the FM906 glass-fibre epoxy specimens GE1 to GE3 compared to the reference (non-cycled) GE0 specimens, see **Figure 8**. In this case the specimens were thermal cycled between 0 and 120°C by external heating and cooling. An increase of 5.5% in ILSS is found after 12000 cycles, which is in good agreement with the previously found 4.8%. [18] Thus, from 12000 to 36000 cycles a further increase in ILSS is found. The increase is expected to be caused by physical ageing of the epoxy. The physical ageing reduces the free volume and molecular mobility of the epoxy. [17,18]

As a result physical ageing stiffens the material. The continuous heating tests at 120°C confirm the positive effect of physical ageing on the interlaminar shear strength. A significant increase in interlaminar shear strength up to 12.2% after 96 hours is found in this case, see **Figure 9**.

The heated FM906 glass-fibre epoxy specimens HGE1 to HGE3 show a decrease in interlaminar shear strength down to -7.3% after 12kC compared to the reference (non-cycled) HGE0 specimens, as can be deduced from **Figure 10**. These specimens were thermal cycled between -25 and 50°C by internal heating and external cooling. The decrease is expected to be caused by internal stress relaxation at the interface of the heater element and the epoxy. The decrease is clearly an effect of the heater element as right beside the heater element (at positions B and C) even slight increases in ILSS values (up to 1.8%) are found compared to the (non-cycled) reference, as can be seen in **Figure 11**.

Specimen HGE4 was thermal cycled between -25 and 72.5°C by internal heating and external cooling and shows less decrease (-5.4% after 12kC) than specimens HGE1 to HGE3 thermal cycled between -25 and 50°C (see **Figure 11**). This is most likely caused by a positive effect of physical ageing, which is dependent on the total heat exposure time and elevated temperature level.

The heated GLARE 5-2/1-0.3 specimens HGL1 to HGL3, which were thermal cycled between -25 and 50°C by internal heating and external cooling, show an initial decrease in ILSS of -5.6% after 4 kC followed by an increase of 1.3% and 2.6% respectively after 8 and 12 kC (compared to the reference (non-cycled) HGE0 specimens). The heated GLARE 5-2/1-0.3 specimens HGL4 to HGL6, thermal cycled between -25 and 72.5°C by internal heating and external cooling, do not show a drop but a steady increase in ILSS (5.7% after 12kC) compared to the reference (non-cycled) HGL0 specimens. No crack or voids or other visible changes were found in the optical microscope images, shown in **Figure 14**, and the drop is expected to be caused by relaxation of the internal stresses. This effect is counteracted by physical ageing at elevated temperature. Thermal cycling for 12000 cycles in -25 and 72.5°C temperature range takes the HGL6 sample 21.7 hours above 70°C as can be seen in **Table 2**. The ILSS results of both the -25 and 50°C and the -25 and 72.5°C temperature range are given in **Figure 12**.

Previous results on heated GLARE 5-2/1-0.3 specimens cycled between -25 and 95°C by internal heating and external cooling showed a decrease in ILSS (down to -7.8% after 12kC) compared to the reference (non-cycled) specimens. [18] It should be noted that in these tests the heater element was s-shaped instead of straight, had 5 mm width instead of the currently used 2.5 mm, and was located at the centre and not at the edge of the samples.

The ILSS test results for the continuously heated GLARE 5-2/1-0.3 specimens AHGL1 to AHGL12 show similar counteracting effects of stress relief and physical ageing. All the ILSS values are decreased after ageing in this case, but continuous heating at 50°C remarkably shows the largest decrease in strength, -37.1% ,after 192 hours, see **Figure 13**. Internal heating to a steady state temperature of 80°C and 120°C shows only a slight further decrease for longer ageing times after the initial drop at 24 hours.

5. Conclusions

The positive effect on the interlaminar shear strength of FM906 glass-fibre epoxy composite after thermal cycling between 0 and 120°C by external heating or continuous heating at 120°C is attributed to physical ageing that stiffens the epoxy matrix.

Internal heating FM906 glass-fibre epoxy samples between -25 and 50°C and -25 and 72.5°C on the contrary showed a decrease in ILSS (-7.3% and -5.4% after 12kC). The decrease is expected to be caused by internal stress relaxation at the interface of the heater element and the epoxy, as right beside the heater element slightly increased ILSS values are found.

After an initial drop (-5.6% after 4kC), expected to be caused by internal stress relief, the GLARE samples cycled between -25 and 50°C show increased ILSS values (up to 2.6% after 12kC). The GLARE samples cycled between -25 and 72.5°C do not show a drop but a steady increase in ILSS (5.7% after 12kC) as a result of physical ageing. Optical microscope images of the heated GLARE samples did not show any cracks, voids or other visible changes due to thermal cycling after 12kC in both temperature ranges.

The ILSS test results for the continuously heated GLARE 5-2/1-0.3 specimens show similar counteracting effects of stress relief and physical ageing and the largest decrease was found at 50°C; -37.1% after 192 hours.

Received: ((will be filled in by the editorial staff))
Revised: ((will be filled in by the editorial staff))
Published online: ((will be filled in by the editorial staff))

References

[1] A. Vlot, J. W. Gunnink, eds, *Fibre metal laminates: An introduction,* Kluwer Academic Publishers, Dordrecht, The Netherlands **2001**.

[2] R. C. Alderliesten, M. Hagenbeek, J. J. Homan, P. A. Hooijmeijer, T. J. De Vries, C. A. J. R. Vermeeren, *Appl. Compos. Mater.* **2003**, 10, 223.

[3] R. C. Alderliesten, *Fatigue and Fracture of Fibre Metal Laminates,* Springer **2017.**

[4] Federal Aviation Administration (FAA). *Airplane and Engine Certification Requirements in Supercooled Large Drop, Mixed Phase and Ice Crystal Icing Conditions*, Federal Register 75 (124) **2010.**

[5] M. Mohseni, A. Amirfazli, *Cold Regions Science and Technology* **2013,** 87, 47.

[6] C. A. J. R. Vermeeren, Th. Beumler, J. L. C. G. De Kanter, O. C. Van der Jagt, B. C. L. Out, *Appl. Compos. Mater.* **2003**, 10, 257.

[7] S. Y. Park, W. J. Choi, H. S. Choi, *Composite Structures* **2010,** 92, 18.

[8] A. A. Costa, D. F. N. R. da Silva, D. N. Travessa, E. C. Botelho, *Materials and Design*, **2012** 42 (1), 434.

[9] H.J. Jiang, L.H. Liang, Y.L. Xiao, J. Chen, Y.J. Xu, X.G. Wang, *Int. J. Heat. Mass. Transfer*, **2018**, 118, 671

[10] H.J. Jiang, X.G. Wang, L.H. Liang, H.L. Dai, *App. Thermal Eng.* **2016**, 106, 161

[11] Fibre Metal Laminates Centre of Competence, http://www.fmlc.nl, acc. 24.3 **2018.**

[12] M. Pacchione, J. Telgkamp, *Proc. ICAS-25* **2006,** 1.

[13] M. Hagenbeek, *Characterisation of Fibre Metal Laminates under Thermo-mechanical Loadings*, Delft University of Technology, Delft, The Netherlands **2005.**

[14] T. Beumler, *Flying Glare,* Delft University of Technology, Delft, The Netherlands **2004.**

[15] A. G. Anisimov, B. Müller, J. Sinke, R. M. Groves, *Proc. SPIE 9435* **2015.**

[16] B. Müller, A. G. Anisimov, J. Sinke, R. M. Groves, *Proc. ETNDT-6* **2015,** 1.

[17] G. M. Odegard, A.Bandyopadhyay, *J. Polym. Sci. Part B* **2011,** 49 (24), 1695.

[18] B. Müller, M. Hagenbeek, J. Sinke, *Composite Materials* **2016**, 152, 106.

[19] B. Müller, S. Teixeira De Freitas, J.Sinke, *Proc. ICCM-20* **2015**, 4212-3, 1.

[20] D. D. L. Wijngaards, E. Cretu, S. H. Kong, R. F. Wolffenbuttel, *Proc. Mod. and Sim. of Microsystems-3* **2000,** 652.

[21] ASTM, *Standard Test Method for Short-Beam Strength of Polymer Matrix Composite Materials and Their Laminates*, D2344/D2344M-13.