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Monitoring chemical degradation of thermally cycled glass-fibre composites using hyperspectral imaging

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ABSTRACT

Nowadays, the application of glass-fibre composites in light-weight structures is growing. Although mechanical characterizations of those structures are commonly performed in testing, chemical changes of materials under stresses have not yet been well documented. In the present work coupon tests and Hyperspectral Imaging (HSI) have been used to categorise possible chemical changes of glass-fibre reinforced polymers (GFRP) which are currently used in the aircraft industry. HSI is a hybrid technique that combines spectroscopy with imaging. It is able to detect chemical degradation of surfaces and has already been successfully applied in a wide range of fields including astronomy, remote sensing, cultural heritage and medical sciences. GFRP specimens were exposed to two different thermal loading conditions. One thermal loading condition was a continuous thermal exposure at 120°C for 24h, 48 h and 96h, i.e. ageing at a constant temperature. The other thermal loading condition was thermal cycling with three different numbers of cycles (4000, 8000, 12000) and two temperature ranges (0°C to 120°C and -25°C to 95°C). The effects of both conditions were measured using both HSI and interlaminar shear (ILSS) tests. No significant changes of the physical properties of the thermally cycled GFRP specimens were detected using interlaminar shear strength tests and optical microscopy. However, when using HIS, differences of the surface conditions were detected. The results showed that the different thermal loading conditions could be successfully clustered in different colours, using the HSI linear unmixing technique. Each different thermal loading condition showed a different chemical degradation level on its surface which was indicated using different colours.

Keywords: thermal cycling, non-destructive testing, aerospace, optical diagnosis, hyperspectral imaging, glass-fibre composites, chemical degradation, ageing, mapping.

1. INTRODUCTION

Thermal loading of glass-fibre reinforced polymers (GFRP) can lead to material degradation. Effects can for example be cracking or delamination due to differences in thermal expansion and corresponding thermal stresses. Molecular changes related to physical ageing can also occur due to exposure at elevated temperatures[1]. When flaw sizes are small or when gradual and global degradation occurs many existing non-destructive testing (NDT) methods like ultrasonic scanning do not capture the material degradation.

Hyperspectral Imaging (HSI) is a hybrid technique that combines reflectance spectroscopy with digital imaging. It was first introduced in astronomy and has a wide range of applications such as, remote sensing, paints and coatings[2], cultural heritage[3,4] and medical sciences[5]. This combination of spectroscopy and imaging results in a three dimensional (3D) space, called a spectral cube, which consists of one spectral and two image dimensions. The benefits from the spectroscopy point of view, is that the 3D cube consists of millions of different measurement points. From the imaging point of view, three colour vision (trichromatic imaging) is extended to hundreds of colour components. Moreover, such a combination enables mapping of areas with similar spectral characteristics.

Currently, the assessment of ageing of materials is performed by destructive testing techniques (eg. ILSS tests). The main disadvantage of such methods is that the structure is partially damaged. Therefore, these techniques are not commonly used for maintenance purposes. Therefore, there is a need for a non-destructive evaluation and inspection method to assess ageing of GFRP materials.

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In this paper Hyperspectral Imaging was used for the assessment and detection of surface degradation due to thermal loading. The capabilities of HSI were tested in two different thermal loading conditions. In the first condition the material was subjected to ageing, i.e. to a constant temperature exposure of 120°C for several hours. In the second condition the material was subjected to thermal cycling, i.e. to periodic temperature changes for several thousand cycles with temperature differences of 120°C. As the exposure of GFRP at elevated temperatures is expected to change the material properties, interlaminar shear strength (ILSS) tests were used to mechanically assess these expected changes. However, ILSS are destructive tests and do not allow the assessment of the chemical degradation. Hence, the HSI was applied for the assessment of the chemical degradation. The test results showed a correlation between the material changes measured with the ILSS tests and the HSI results. Thus, this study shows that HSI was sensitive enough to measure the effect of chemical ageing on the GFRP surfaces.

2. SPECIMENS

2.1 Material

The material used in the tests was a glass-fibre epoxy composite laminate with $[0/90]_{4s}$ layup. This laminate was manufactured in the autoclave using unidirectional FM906 glass-fibre epoxy prepreg layers, which was cured at a temperature of 180 °C and a pressure of 6 bar[6]. The same prepreg is used in the aerospace industry to manufacture high strength GLARE (HSS GLARE). This special Fibre Metal Laminate (FML) combines unidirectional FM906 prepreg layers together with thin 7075-T6 aluminium sheets to improve strength and temperature resistance over standard Glare[6]. Another special GLARE grade, designated heated GLARE and based on the same prepreg, integrates heater elements into the laminate for de-icing and/or anti-icing purposes[7]. These special GLARE grades have been developed for elevated temperature applications up to 120 °C. In the current research the possible effects of long-term exposure at elevated temperatures are investigated.

Long-term exposure at elevated temperatures below the glass transition temperature for extended periods of time (annealing) can lead to changes in material properties. The material can undergo an increase in density (volumetric relaxation) and/or a decrease in molecular configurational energy (enthalpy relaxation) of amorphous or semi-crystalline materials. This is called physical ageing. An extensive overview of epoxy ageing is given by Odegard and Bandyopadhyay[1]. They also list a number of studies that have been conducted on the physical ageing of fibre reinforced epoxy composites. These studies indicate that the ageing response of the composite material is very similar to pure epoxy resin. Moreover, physical ageing has also little influence on the fibre-epoxy interface strength and does not alter the load-transfer characteristics of fibre reinforced composites as shown by Truong and Ennis[8].

The effect of ageing on the mechanical properties of epoxy is summarized in Table 1. It can be seen that epoxy shows embrittlement and hardening as a result of physical ageing. While the modulus, shear and compression yield are positively affected, the tensile strength, fracture toughness and strain to failure decrease. For the glass-epoxy layers in heated GLARE similar effects are expected when long-term and/or repeated exposure to elevated temperature occurs. In glass-fibre epoxy physical ageing is most apparent in matrix dominated properties. Shear and creep properties will see improvement, but fatigue crack initiation will be much faster. For GLARE laminates a small decrease in the epoxy tensile strength is expected to have limited effect on the overall properties, since the stiffness of and strength properties of the aluminium layers and the glass-fibres are several times higher than those of the epoxy[9].

Table 1 General effects of ageing on the mechanical properties of epoxies, derived from Odegard and Bandyopadhyay[1].

Mechanical property	Effect	Description of the found effect
Elastic modulus	⇧	no or modest increase
Compression yield	⇧	up to 15% increase observed
Shear	⇧	similar trends as in compression
Hardness	⇧	embrittlement
Tensile strength	⇩	growth of microcracks at lower loads
Strain to failure	⇩	decreases observed
Fracture toughness	⇩	critical energy release rate for initial crack growth (G_i) dramatically reduced

2.2 Dimensions & Thermal loading

To investigate the effect of ageing on the FM906 glass-fibre epoxy composite two approaches were followed. In the first approach 50 mm x 100 mm x 2 mm samples were cut and the interlaminar shear strength (ILSS) specimens were subjected to 120 °C for 24, 48 or 96 hours in the oven prior to testing. Table 2 gives the test matrix with the thermal loading conditions. The ageing times were chosen in order to match with the exposure times of the thermal cycled specimens. Assuming that the effect of ageing is limited below 70 °C the total exposure was in the same range for both the specimens that have been thermal cycled and the ones that were exposed (aged) continuously to elevated temperature in the oven. After ageing, five ILSS specimens were tested from each ageing time batch and compared with the results from the reference (non-aged) batch. The interlaminar shear strengths of the specimens were tested in a three-point bending setup[10]. The specimen dimensions were 4 mm x 20 mm x 2 mm (width x length x thickness). Figure 1 depicts the sample and specimen dimensions.

Table 2 Overview of the specimens and different thermal loading conditions.

Nomenclature	Thermal loading	No. of cycles
Reference	Non-exposed after manufacturing	-
Aged 24	Continuous exposure at 120 °C for 24 h	1
Aged 48	Continuous exposure at 120 °C for 48 h	1
Aged 96	Continuous exposure at 120 °C for 96 h	1
0/+120 4000 cyc.	Thermal cycling between 0 °C and 120 °C	4000
0/+120 8000 cyc.	Thermal cycling between 0 °C and 120 °C	8000
0/+120 12000 cyc.	Thermal cycling between 0 °C and 120 °C	12000
-25/+95 4000 cyc.	Thermal cycling between -25 °C and 95 °C	4000
-25/+95 8000 cyc.	Thermal cycling between -25 °C and 95 °C	8000
-25/+95 12000 cyc.	Thermal cycling between -25 °C and 95 °C	12000

In the second approach 50 mm x 100 mm x 2 mm samples were thermally cycled both between 0 °C to 120 °C and -25 °C to 95 °C for 4000, 8000, and 12000 cycles (cf. Figure 1). The thermal cycling tests were conducted using a specifically designed thermal cycling setup which uses Peltier elements for cooling and/or heating[11,12]. Thus, the samples were exposed in total approximately 24, 48, and 72 hours above 70 °C as regards to the 0 °C to 120 °C after 4000, 8000, and 12000 thermal cycles respectively. For the -25 °C to 95 °C thermal cycling, the exposed hours above 70 °C were 32, 64, and 96 hours after 4000, 8000, and 12000 thermal cycles respectively. Each thermal cycle duration was 60 sec and 95 sec for the 0 °C to 120 °C and for the -25 °C to 95 °C thermal cycles respectively. After thermal cycling, five ILSS specimens were cut from each sample, were tested and the results were compared to the non-cycled reference specimens.

The thermally cycled material and its corresponding reference specimens come from a different autoclave batch than the aged material and reference specimens. Thus, a separate reference sample was taken from each plate.

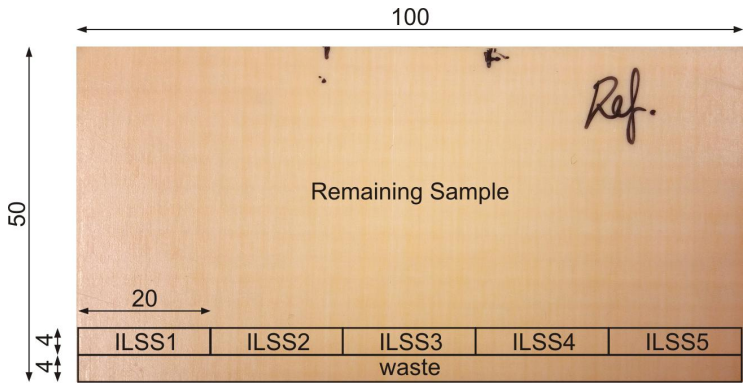


Figure 1. Photo of the reference sample including the positions of the ILSS specimens, units are in Millimetres.

All specimens were scanned before and after thermal loading using Hyperspectral Imaging and compared to the non-exposed specimens to investigate if a correlation between the imaging technique and the amount of thermal loading can be established. Furthermore, all of the exposed specimens were scanned before and after thermal loading using an ultrasonic C-scan device. There were no delaminations, cracks or other defects detected with the C-scan technique.

3. HYPERSPECTRAL IMAGING METHOD

3.1 The HSI setup

The Aerospace Non-Destructive Testing Laboratory at TU Delft is equipped with an imaging monochromator IMSPECTOR V10E (Specim©), which has a spatial resolution of 1300 pixels and operates between the 400 nm - 1000 nm wavelength range with a bandwidth of 2.8 nm. In Figure 2 a photo of the setup is presented. Acquisition was achieved through Specim's default SpectralDAQ solution software©, that allowed for the control of basic sensor parameters (frame rate [fps], exposure time [ms], spectral and spatial binning). Scanning of the specimens was performed with an automated scanning platform. The objective lens used has a focal length of 35mm that enabled imaging of approximately a 100 mm of field of view. An additional flattening filter (Specim©) was used in front of the objective lens to enhance the information obtained at the edges of the sensors sensitivity range (UV and IR part of the spectrum). To avoid any specular reflections, the light sources (3 tungsten 30W Halogen) had a 45 degree angle of incidence with respect to the sample and the sensor. This ensured that only diffuse scattering caused by the surface roughness and scattering centres underneath the surface was recorded by the camera.

3.2 Calibration

Calibration of the system was achieved by a Diffuse Reflectance Target (DRT) made out of spectralon (Laser2000). The target was used after the measurements with the same conditions to allow further normalization of the data. Normalization was used to flatten the spectral curves, and to correct for the interference noise coming from the SPECIM optics and the light inhomogeneities. Lastly, dark current was corrected by subtracting images acquired with the camera shutter closed while keeping the sensor shutter time constant.

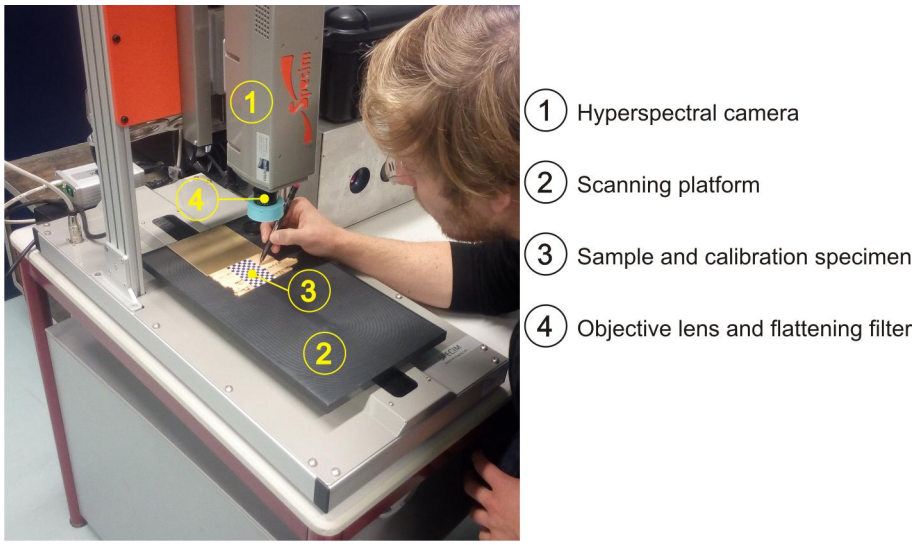


Figure 2. Photo of the hyperspectral imaging setup.

3.3 Measurements and Analysis

Two sets of measurements were performed. In the first acquisition the aged specimens which were exposed to a constant temperature of 120°C and the reference specimens were measured. In the second acquisition the specimens which were thermal cycled and the reference specimens were measured. Before each of those acquisitions, the diffuse reflectance target was used for normalization purposes.

Analysis of the HSI data was achieved with a tailor made software analysis platform (TIPP, TU Delft) specifically developed in the Aerospace Non-Destructive Testing Laboratory at the Delft University of Technology[13]. Reference spectra were initially extracted from 5 points in each sample and then averaged to reduce noise. This resulted in six reference spectrums, one for each sample category (three different numbers of cycles and two temperature ranges). These reference spectrums were then used for analysis with a standard linear unmixing algorithm. This algorithm produced separate image maps with the weight of each reference sample contribution to the overall spectrum. Thus, a specific pseudo-colour was assigned to each image map before merging all image maps to a final image. The results allowed imaging with high contrast of the surface chemical degradation and the surface areas.

4. EXPERIMENTAL RESULTS

4.1 Aged specimens

Differences in the chemical degradation of the surface are shown in Figure 3. The three different ageing times under constant temperature were compared with the reference specimen which was not exposed to any thermal loading after manufacturing. The reference specimen was kept at a room temperatures of approximately 22 °C. The effects of the different thermal loading conditions (different thermal exposure times) were mapped using different colours.

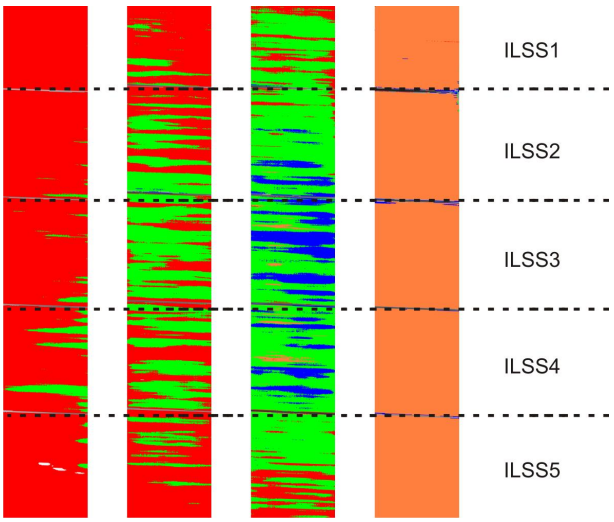
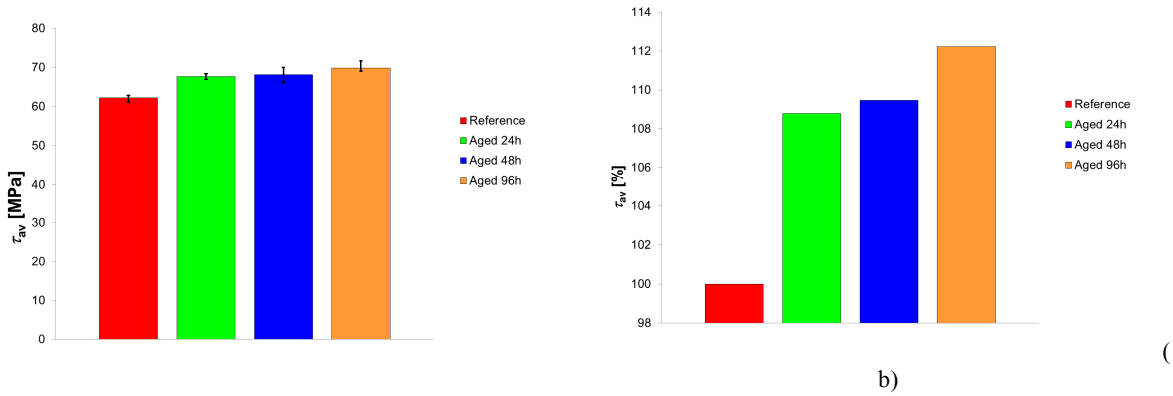


Figure 3. Hyperspectral images of the aged ILSS specimens: (a) Reference, (b) Aged 24h, (c) Aged 48 h and (d) Aged 96 h.

The reference (non-loaded) specimen was mapped in red while the effects of the chemical degradation of the aged specimens were mapped with green (early stage), blue (higher degradation), and with orange (highest degradation measured). The colour shift from the reference specimen to the specimen with the longest thermal exposure, presented a strong relationship with the thermal exposure times. After 96 h of a constant exposure at a temperature of 120°C, the HSI result of those specimens showed a homogeneous image map.

Figure 4 (a) compares the absolute ILSS values of the glass fibre-epoxy (FM906) samples without (Reference) and with a continuous exposure at 120°C (Aged) for 24 h, 48h and 96 h. The reference ILSS value was 62 MPa. Figure 4 (b) shows that ageing for 24 h, 48 h and 96 h increased the ILSS about 9 %, 10 % and 12 % respectively.

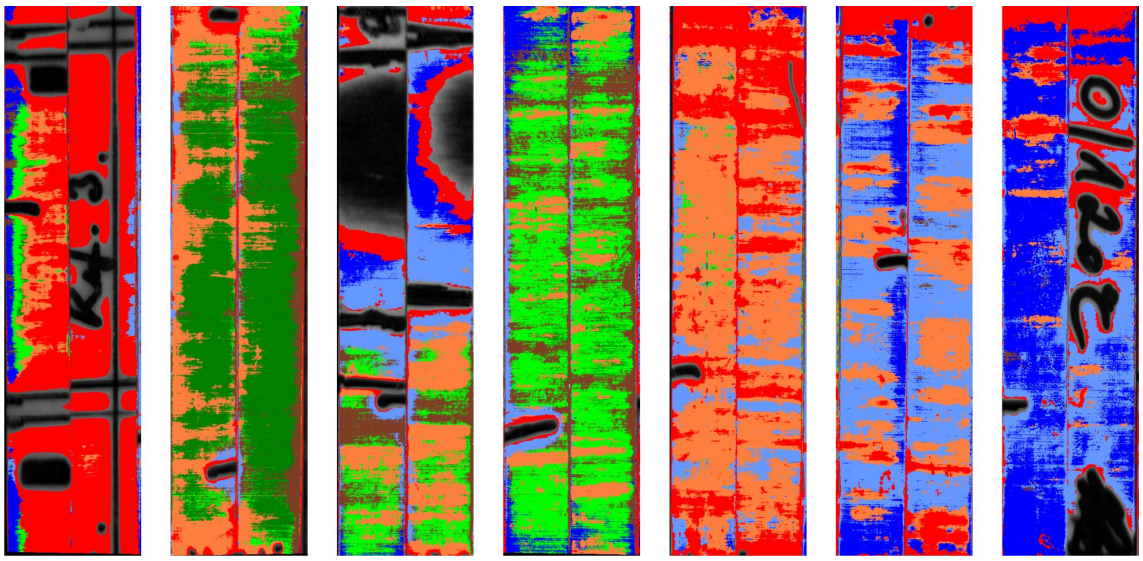


(a) b)

Figure 4. ILSS results of the aged specimens: (a) Absolute values and (b) percentages compared to the reference.

4.2 Thermally cycled specimens

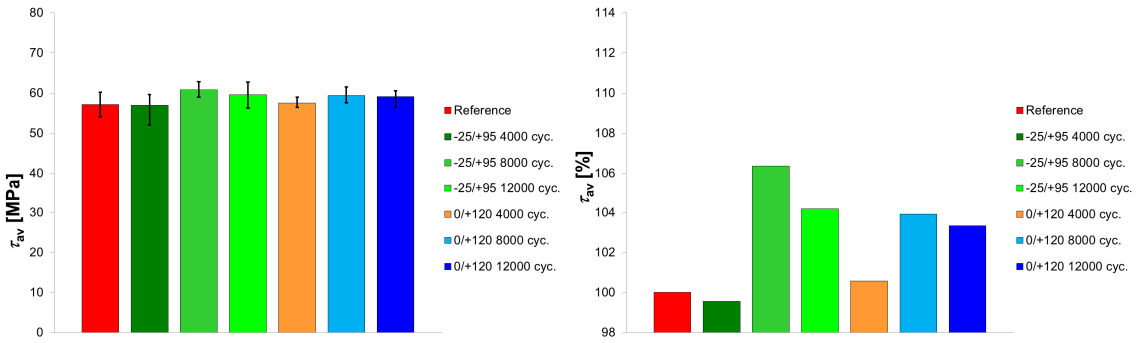
The differences in the chemical degradation of the thermal cycled specimens surface are shown in Figure 5. The reference sample was kept at a room temperatures of approximately 22 °C (red colour). The thermally loaded specimens presented a more complicated chemical degradation map on their surfaces but showed a clear relationship between the different number of cycles and the different category colours. The chemical degradation of the samples which were exposed to 4000, 8000 and 12000 cycles at the lower temperature range (from -25 °C to 95 °C) was presented with a colour shift from orange, to dark green, and light green, while the chemical degradation of the samples which were exposed to 4000, 8000 and 12000 cycles at the higher temperature range (from 0 °C to 120 °C) was mapped with a colour shift from orange to light blue, and to dark blue.



(a) (b) (c) (d) (e) (f) (g)

Figure 5. Hyperspectral images of the thermal cycled specimens: (a) Reference, (b) -25/+95 4000 cycles, (c) -25/+95 8000 cycles, (d) -25/+95 12000 cycles, (e) 96 h 0/+120 4000 cycles, (f) 96 h 0/+120 8000 cycles and (g) 96 h 0/+120 12000 cycles. Note that black and grey areas are not categorized with the algorithm.

Figure 6 (a) compares the absolute ILSS values of the glass fibre-epoxy (FM906) samples not-cycled (Reference) and the thermal cycled specimens with 4000, 8000 and 12000 cycles in the two different ranges (-25 °C to 95 °C and 0 °C to 120 °C). The reference ILSS value was 57 MPa. Figure 6 (b) shows that, except from one value, the ILSS values increased due to thermal cycling. The maximum increase was about 6 % after 8000 cycles[12].



(a) (b)

Figure 6. ILSS results of the thermal cycled specimens: (a) Absolute values, (b) percentage compared to the reference.

5. DISCUSSION

In this paper the application of hyperspectral imaging for the detection of surface degradation of glass-fibre reinforced polymer samples after ageing is demonstrated. The results are promising and allow a qualitative assessment of the surface degradation by comparing the thermally loaded samples with a reference (un-loaded) sample. The colour shifts and the ILSS values of the specimens correlate. However, at this stage of the research a quantitative assessment of the material degradation using HSI was not possible as more tests and further in detail investigation about the correlation of the colour shift rate (different colours) and the amount of ageing are required.

Particularly, in the constant temperature aged specimen test, a clear relationship between the number of ageing hours and the ILSS strength test values was found. Since, in the case of a constant temperature ageing process, i.e. when comparing Figures 3 (different colours) and 4 (different ILSS values) a strong relation between the hyperspectral imaging and ILSS test results was found. Both the colour shift and the increase of the ILSS values continuously changed with increasing exposure times. Thus, the HSI technique showed that the surface degradation, the ILSS values and the amount of ageing (exposure times) were following a similar trend.

Specimens that were thermally cycled showed a more complex thermal loading behaviour which resulted in a less clear relationship between the number of cycles and the ILSS strength tests. In contrast to this, HSI tests presented a clear trend of the surface degradation and the number of cycles. However, when comparing Figures 5 (different colours) and 6 (different ILSS values) a relation between the hyperspectral imaging and ILSS test results is visible. Although, the relation was not that clear as when comparing the Figures 3 and 4, a correlation between the colour shift and the change of the ILSS values was found. Hence, the effect of the more complex thermal loading conditions on the HSI results and the ILSS test results was shown, but was more complex to map compared to the case of the constant temperature exposure (ageing process).

When comparing the differences between the two temperature ranges, differences were visible but they not clearly related to each other. It must be noted that one specimen was burned in one small area (black), as shown in Figure 5c. Based on this clear colour change of the burned area, it can be understood that the higher temperatures which caused this local burning reflect a higher chemical degradation due to the temperature exposure. Since, around the burned area bluish colours were mapped, the following conclusion can be drawn: The higher temperature range (0 °C -120 °C) caused a higher chemical degradation. Although, this trend can be understood, it requires more study to further characterise the ageing differences due to the different levels of chemical degradation caused by the different thermal cycling ranges and number of cycles.

Furthermore, it needs to be stated clearly that the colour schemes cannot yet be modified with the current software. Thus, the colour schemes used for the examination of the aged and thermal cycled specimens were different and the results presented in the Figures 3 and 5 can therefore not be directly compared. Homogenising the colour mapping for the different thermal loading conditions in order to directly compare the results is a topic for future research.

6. CONCLUSIONS

FM906 glass-fibre epoxy composite has been thermally loaded by means of thermal cycling and continuous temperature exposure. The thermal cycling and continuous exposure showed increased ILSS values, up to 6 % and 12 % respectively. The changes of the ILSS values are likely to result from changes in the microstructure, as microscopic investigations and ultrasonic C-scan results after thermal cycling showed no changes in the physical structure such as cracks or delaminations.

Using Hyperspectral Imaging, differences of the surface conditions were detected. The results showed that the different thermal loading conditions could be successfully clustered using the HSI. Each different thermal loading condition showed a specific wave length spectrum which were indicated using different colours.

As a final conclusion we were able to qualitatively assess ageing of FM906 sample, but for a quantified analysis further studies have to be performed.

ACKNOWLEDGMENTS

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