Pre- and post-bond inspection for assuring quality of adhesive bonds of composites after thermal treatment

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Abstract
The research presented here is focused on non-destructive assessment of CFRP structural parts. The performance of adhesive bonds depends on the properties of the surfaces that are joint. Improper preparation of the surface may lead to weak bond that cannot carry the desired load. This is the reason why there is a search for methods of surface assessment before bonding. Moreover, it is important to have reliable techniques that allow to verify the integrity of the adhesive bond after manufacturing or bonded repair. The structural joints should ensure safe usage of a structure. In reported research we focus on the laser induced fluorescence (LIF) method and the scanning laser Doppler vibrometry (SLDV) for assessing of the surface state. In the context of adhesive bond assessment the electromechanical impedance (EMI) method is studied. The LIF is full non-contact measurement method. The SLDV uses a surface bonded piezoelectric sensors for excitation and laser beam for sensing of the elastic waves. Similar sensor is used for measuring spectra in the EMI method. The investigated samples were made of CFRP (Carbon Fibre Reinforced Polymer) layers. The CFRP plates were treated at elevated temperatures. The influence of this temperature was studied using LIF and SLDV. The thermally treated plates were bonded to untreated plate and then they were studied with the EMI method to assess the bonding quality.

Keywords: Adhesive joints, CFRP, Composite laminates, Composite materials and repairs, Composites, electromechanical impedance, NDT

1. Introduction
The research presented here is focused on non-destructive assessment of surface prepared for bonding and the bonded joint. Wrong preparation of the surface may lead to weak bond. The promising methods for surface assessment are, for example, the electronic nose [1] and the aerosol wetting technique [2]. Apart from the problem of surface assessment the another important subject is to assess the bonded part. The examples of techniques investigated for this task are electromechanical impedance [3] and laser-induced shock waves [4]. The research reported here aims to non-destructively assess the surface of samples that were thermally treated at elevated temperatures. The second aim is to inspect the adhesive bond of samples one of which is after this thermal treatment. The pre-bond thermal treatment results reported in the literature shows that the mode-I energy release rate (GIC) value decreases by 10 and 13 % for treatment at 190 and 200°C, respectively. In the case of treatment at 210°C an increase by 9% was observed [5].

The intention of this paper is to study the laser induced fluorescence (LIF) and laser vibrometry (SLDV) methods method for assessment of the surface condition. The adhesive bonding quality was investigated by the electromechanical impedance (EMI) method.

The investigated samples were made of Hexcel M21E/IMA material. The samples for surface assessment were made with 8 plies and layup sequence: [0, 0, 45, -45]s. Their size was 200 x 200 mm². The samples for adhesive bonds assessment comprised of two plates with the same layup sequence adhesively bonded together. The plates were bonded with FM 300-2 adhesive cured at 121°C. The bonded sample size was 100 x 100 mm².

In total 24 samples were investigated. Twelve samples for surface assessment comprised of three reference samples (RREx) and 9 thermally treated samples. The thermally treated samples
were hold in air circulation oven for 2h. Three levels of treatment were investigated, so three samples were treated at 220°C (RTD1x), three at 260°C (RTD2x) and three at 280°C (RTD3x). The same scheme was followed for twelve adhesively bonded samples. The bonded reference samples were comprised of two plate without any thermal treatment. The modified bonded samples comprised of one plate after thermal treatment and one without any treatment.

2. **Surface assessment using SLDV**

In vibrometry technique propagating elastic waves are used to investigate the state of an object. Such waves in relatively thin plates are called Lamb waves, plate waves or guided waves. Propagating elastic waves can be influenced by discontinuities (cracks, delaminations, etc.), thickness change or variation of material properties as elastic modulus or density. In reported research the waves were excited in the samples (flat, 200 x 200 mm²) by a piezoelectric transducer (10 mm disc) that was glued to the sample surface at one corner. The excitation signal was in a form of a pulse in order to ensure excitation of waves in broad frequency band. The wave sensing was realised by Polytec SLDV. The laser measurements were conducted in points defined along the sample diagonal. In this way it was possible to register the propagating wave field less affected by the boundaries, and then process it to search for anomalies related to the sample surface state.

2.1 **Wavenumber processing**

The time signals registered along the sample diagonal were processed in order to transform the signal from time-space domain (t-s) to frequency-wavenumber space (f-k) domain. For this purpose a two dimensional Fourier Transform was used. This transformation was preceded by an energy compensation in space. In result a curve presented at Figure 1a was obtained. Such curve is called a dispersion curve and is characteristic for a sample made of given material with given thickness. Next, a thresholding procedure was applied in order to create a binary image of the dispersion curve (Figure 1b). For the data in Figure 1b linear fitting was conducted. Inverting the value of slope of the line gives a linear approximation of the elastic wave group velocity \( c \). This velocity was taken for comparison of the investigated samples.

![Figure 1: Result of transformation from time-space into frequency-wavenumber domain; a) – result of the transformation, b) binary image of the curve](image)

2.2 **Results**

The estimated group velocities values were plotted in Figure 2. There are clear differences between the three reference samples. The highest value for reference samples was obtained for RRE3 sample. The lowest value is observed for RRE1 sample. Looking at the results of thermally treated sample the highest values were obtained for the RTD11 and RTD13 samples.
These are the samples treated at the lowest temperature (220˚C). The highest and lowest velocity value obtained for reference samples define an interval. Most of the results fall in this interval. Only the values for following samples are above the defined interval: RTD11, RTD13, RTD21.

Figure 2: Calculated wave group velocity for three reference samples (RRE1x) and nine samples after thermal treatment

3. Surface assessment using LIF

In Laser Induced Fluorescence (LIF) technique spectra were recorded using laboratory system equipped with excitation source - Nd:YAG, 6 ns pulse, 1064 nm laser (Brilliant B, Quantel) equipped with harmonic generation modules: SHG – 532 nm, THG – 355 nm and FHG – 266 nm. The detection of fluorescence were carried on using 0.3 m monochromator (Andor 303i) equipped with 600 g/mm grating and ICCD detector (Andor Istar). Fluorescence signal from analyzed surface was collected using quartz lens and focused on the entrance of the optical fiber connected to the monochromator entrance slit. LIF spectra were recorded in the time integration mode. The minimal time gate of the ICCD detector was 5 ns. The LIF spectra were recorded for laser excitation at 532 nm. The emission spectra were recorded in 10 independent points on sample surface and averaged.

3.1 Results

The results of LIF measurements were plotted in Figure 3. There is high intensity for thermal degradation at the first level (RTD1x) observed. This case was achieved by keeping the samples at 220˚C in an oven. However, the samples RTD2x and RTD3x were kept in higher temperatures, there is no correlation of the intensity with the used temperature. In previously published work [6] we saw good detection of TD scenario with the LIF intensity measurement at 532 nm excitation. Different levels of thermal treatment correlated with the intensity. The intensity value was growing with the growing temperature of treatment. Here we see different behavior. The first important difference is that the new samples (RTD) were ground down to the fibers. The samples investigated in [5] were measured just after the thermal treatment without any surface. It is possible that layer responsible for fluorescence was removed by grinding and the intensity increase is not observed any more. The second important difference is that the RTD samples were treated at higher temperatures than before. What is important the other side of the sample was not prepared/modified in any way. It was decided to measure this second side and compare the results with the previously obtained. The new result is presented
in Figure 4. Again the highest intensity is observed for RTD1x samples. The RTD2x and RTD3x samples have intensity values on the level of reference samples. There is also a clear difference in vertical scale between Figure 7 and Figure 8. This rather should be related to the considerable time interval between these two measurements. Recalling the previously published results [6] there was a clear intensity increase up to 210°C. Now also an increase is observed but only for 220°C.

Knowing that the behaviour of both sides of the thermally treated samples are similar for 532 nm excitation, a new approach was proposed. Different surface excitation was tested. The investigation for thermally treated samples was repeated using ultraviolet (UV) excitation. The
source of excitation available at the laboratory was 370 nm. It was observed that for this UV excitation the reference samples have higher intensity than the thermally treated samples (Figure 5). Neither of the thermal degradation levels were detected.

Figure 5: LIF Results for reference (RREx) and thermally treated (RTD) samples measured in UV range

4. Bond assessment using EMI

The EMI method is considered as one of the NDT or SHM methods. The method principle is based on a piezoelectric sensor that is bonded to the inspected structure. During the measurement, electric quantities of the sensor are gathered for selected frequency band. The examples of investigated quantities are resistance, conductance, reactance or absolute value of impedance. Due to direct and converse piezoelectric effect, the sensor excites the structure and senses the response from it. This electromechanical coupling causes the registered impedance spectra to be modified by the presence of the host structure. Various structural factors have its influence on the registered spectra. Appearance of additional resonance peaks, peak shift in frequency or magnitude change can be treated as indicator of defect of the structure. In order to extract damage related features for the EMI spectra various frequency bands are analyzed. These bands depend on the inspected structure, the used piezoelectric sensor and the abilities of the available equipment. In this particular work we propose to focus on wide frequency band from 3 to 5 MHz. The band was selected as symmetric around the thickness resonance frequency of the used transducer. In the present work admittance (Y) was investigated on a complex plane.

The admittance characteristic on complex plane comprise of G (conductance) on the horizontal axis (real) and the B (susceptance) on the vertical axis (imaginary). The proposed comparison index is based on Frechet distance (FD). FD is the minimum distance required to connect two points constrained on two separate paths, as the points travel without backtracking along their respective curves from one endpoint to the other [7]. The definition is symmetric with respect to the two curves. The promising results of using FD to assessment of adhesive bonds contaminated with release agent was presented in [8].
4.1 Results

The results of calculating of the FD value is depicted in Figure 6. The result is in the matrix form. It is symmetrical along the diagonal. As can be noticed the results for not all measured samples are depicted here. The reason for rejecting of some of the samples was due to some anomalies in the registered spectra. They could be related to improper bonding of the piezoelectric sensors. The main observation from the obtained matrix is that there is a clearer difference between samples RTD31 and RTD32 and the rest of the samples. These two samples were treated with the highest temperature (280°C). The differences between the rest of the samples are not so evident. The exception is the RTD11 sample.

![Figure 6: Frechet distance calculated for the reference and thermally treated samples](image)

![Figure 7: Frechet distance calculated for all samples in relation to RRE1 sample](image)
In order to facilitate the result interpretation one row of the matrix was plotted in Figure 7. This result depicted the FD value calculated in relation to RRE1 sample. The difference between reference samples is really low. The values for the samples with 280°C treatment significantly differ from the rest. It should be also noticed that the highest value is observed for RTD31 sample. According to the ultrasonic testing results, this sample has a delamination caused by the heating [9] before bonding. This high value could be the indication of this heat-induced damage.

5. Conclusions

Two techniques were used to assess the thermal treatment of the CFRP samples. The LIF technique showed clear difference for samples treated at 220°C. It was observed for excitation at 532 nm. What is important the detection was possible on both faces of the sample (after grinding and not ground). It seems that 260 and 280°C temperature have different influence on the sample so LIF intensity is not influenced. The detection using different wavelength (370 nm) was not possible. Using SLDV also differences for samples treated at lower temperatures were observed. Two samples treated at 220°C, and one at 260°C were detected. Assessment of the bonded samples with EMI allowed for clear separation of the samples treated at the highest considered temperature (280°C).

The research is continued with different types of pre-bond surface modifications.

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References
