



HOT-WET CONDITIONING AND THE SENSITIVITY OF MECHANICAL TESTING WITHIN SURFACE TREATMENT QUALIFICATION FOR AIRCRAFT

D. Blanco Garde^{*,1}, F. Mews^{*,1}, J. Jokinen¹, N. Pournoori¹ & M. Kanerva^{1**,1}

¹Materials Science and Environmental Engineering, Tampere University, Tampere, Finland

* Authors contributed equally on producing the work

**Corresponding author

Abstract

The verification of high-quality surface treatment on composite aircraft structures is not a standardized action and the relationship between certain surface quality measures and mechanical strength is challenging to determine. Surface treatments are necessary for durable adhesive bonds either within aircraft assembly or repair bonding during service engineering. This study focuses on comparing effects of environment (ambient versus hot-wet) on adhesive bond strength when carbon fibre-reinforced composite surface has been qualified by contact angle measurements. Along with the experimental results of this work, a survey about the state-of-the-art of surface quality determination is also given. The results indicate that specimen conditioning is needed to reveal lowered adhesion for certain surface contamination, such as grease on fresh-roughened surfaces.

Keywords: adhesive bonding, surface treatments, quality assurance, composite materials

1. Introduction

Adhesively bonded repair patches are the most potential technique of carrying out repairs of primary structures not only in military vehicles but in civil composite airliners in future. However, great amounts of research are needed to determine reliability levels for certain surface treatments, bonding processes, damage tolerance with cracks, and crack halting capabilities. Overall, successful and confirmed surface treatments on composite structures are of prime importance to reach further performance, e.g., in terms of mechanical durability. This study is about the research of methods to confirm certain surface treatment quality. For metal structures, there are rather established processes, e.g., those of applying roughness and contact angle measurements in confirmation of successful treatment (e.g. [1]). For composite structures, the inherent variation of elemental composition due to different constituents challenges the techniques that rely on even surface response in time and relatively small amount of measuring points. It is important to note that not only static strength is required of adhesive bonds in aerospace structures, but also appropriate long-term durability is necessary in harsh environments [2][3].

Surface quality of composite laminates for adhesive bonding has been determined in the current scientific literature by using contact angle measurements, roughness measurements, infrared Fourier transform spectroscopy (FT-IR), scanning electron microscopy, and X-ray photoelectron spectroscopy (XPS) [4][5][6][7]. For adhesive bonding, not only roughness but chemical composition affects – in terms of surface energy and chemical inertness – the adhesion formation within the bonding process. Contact angle (CA) measurements have been used in a systematic way to confirm surface quality and surface energy components on metal surfaces, but the method has several drawbacks on composite surfaces. Directional surfaces, due to unidirectional fibre reinforcement, tend to form non-sessile droplets and challenge the determination of single representative CA values.

Composites have inherent variation of elemental composition due to matrix and reinforcement and the local variation can affect the surface energy as well as the droplet shape and determined CA [8]. New techniques, such as CA determination based on imaging above the droplet have been developed to mitigate droplet shape discrepancies [9].

In this work, the specimen conditioning is studied to eventually compose a qualification process for surface treatments of composite structures and repairs. Aerospace grade carbon fibre reinforced plastics (CFRP) are used as the material of joints. For a valid qualification process, its methods must offer high reliability for each stage (see schematic in Figure 1). The method to correlate mechanical strength with certain surface character must be sensitive to distinguish failed surface treatments, e.g., due to contamination. Additionally, the most suitable test specimen condition for the characterisation tests needs to be defined.

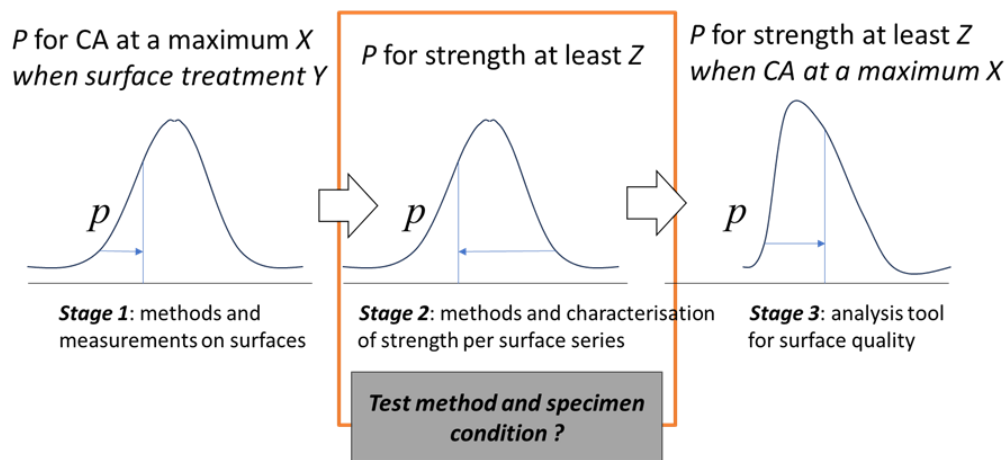


Figure 1 – Schematic of a workflow for creating an evaluation tool for composite surface quality on a reliability basis with probabilities P (P based on respective probability density functions p).

2. Methods

2.1 Materials and specimen preparation

In this research work, we study aerospace grade composite (AS4 carbon fibre and 3501-6 epoxy matrix, forming the CFRP prepreg) as the adherend surface. Different surface conditions are applied to test for the robustness of surface quality validation (baseline roughening and contamination by grease). The laminates of this study included the lay-up: [0/0/0/45/-45/0/0/45/-45/0]S to prepare single-lap shear (SLS) test specimens and double cantilever beam (DCB) fracture test specimens.

The quality of the treated surfaces is characterized by using a hand-held contact angle measuring device Surface Analyst™ SA3001 (Brighton Science (earlier BTG)). This device is capable of industrial measurements, e.g., performed on a wing or fuselage inside hangars or even as (open-air) field measurements. Surface Analyst performs upwards measurements of H₂O droplets, supplied automatically from a standard water cartridge (provided by Brighton Science) of purified water. In addition, the droplet application by this device uses the ballistic application. The surface treatments covered so-called baseline (BL) roughening of the surface by random direction movement and use of P180 grit sanding paper (Mirka). A contamination study (GR) was carried out using aerospace grade grease (GN 22, NYCO); grease was applied in excess amount of the bond surfaces after BL roughening and wiped carefully (to remove excess grease, by using clean cotton cloth).

After the treatments, structural joints (of secondary bonding) are prepared using epoxy B-stage adhesive film (FM 300-2, Solvay), vacuum bagging, and oven cure (120 °C). The temperature ramp of the oven was 2 °C/min and (vacuum) pressure difference of 0.7 bar was applied. The dwell time was two hours (Figure 2).

The mechanical strength related to each surface test series (BL or GR) and specific CA was determined using the SLS testing by using specimens with a total length of 11" and width of 1" (overlap length 0.5"). Additionally, DCB testing was applied to GR series to trial the use of fracture testing as a surface quality control method. DCB testing here followed the advises in the ISO 25217:2009 standard.

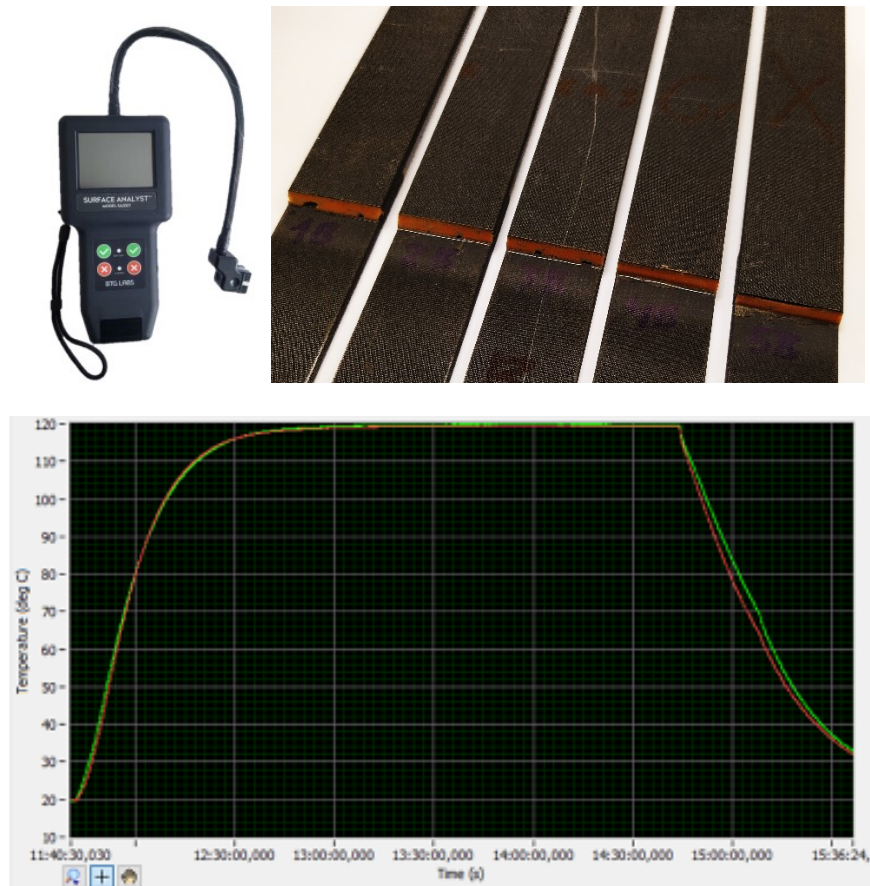


Figure 2 – The CA measurement device for industrial use, the test specimen geometry (with adhesive fillets controlled) of the SLS series, and an example of temperature profile of the cure process (re-printed from [10]) as applied in this work.

2.2 Environmental conditioning

Environmental conditioning can affect adhesion at glue-composite interfaces, thus, improves the sensitivity of a mechanical testing method to surface treatment variation. In this work, the effect of hot-wet conditions was studied in two ways: 1) conditioning of test joints in a chamber for 15 days at an elevated temperature (60 °C) and moisture (90-95% RH) (i.e., ETW conditioning); 2) appending of conditioning (in addition to ETW) with purified water immersion at an elevated temperature (60 °C) for 27 days (i.e., ETWi). After conditioning, the specimens were brought directly to the test machine and immediately tested moist and 'warm' (surface temperatures were qualitatively checked during testing by using infrared thermal camera).

3. Results

3.1 Contact angle range identification (RI) per surface treatment and contamination

The two studied surface treatments (BL, GR) result in very different contact angles, as expected. BL treatment represent typical value of water CA on properly roughened CFRP surface, in the range of 26-46° (CA by Surface Analyst). After contamination with grease (GR), the contact angles on different size of laminates were 65-79° (CA by Surface Analyst). Very detailed analysis of these CA measurements can be found in two academic theses [8] [10]. For example, the BL surface treatment was analyzed in terms of the surface quality. In this, the repeatability of CA measurements on many locations on surface treated CFRP plates were studied. Also, the aim was to evaluate if the distribution of CA is normal or not (via the Q-Q plot and Chi-square goodness (χ^2) fit). The results of the CA measurements and the analysis about the distribution of CA values on CFRP plates are shown in Figure 3. It was noted that the distribution was not exactly normal (per Q-Q plot and respective hypothesis rejection for χ^2 fit).

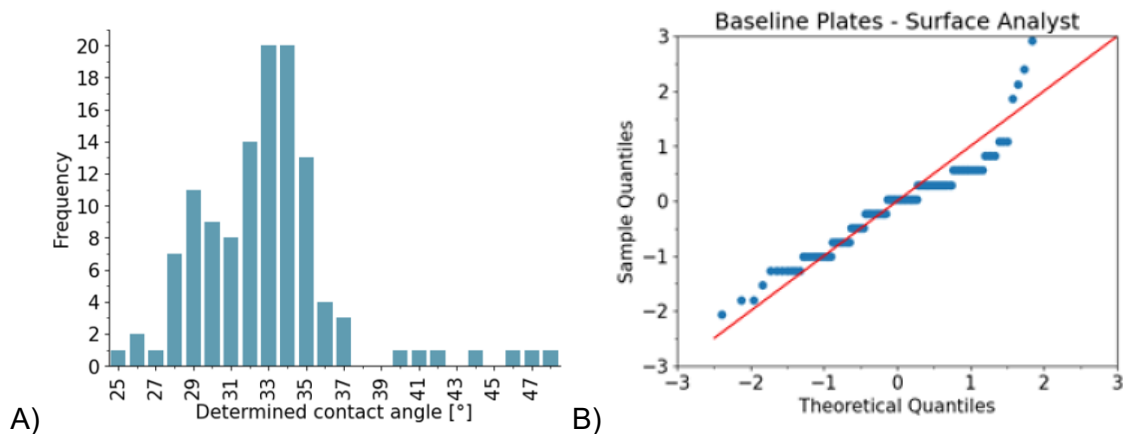


Figure 3 – CA measurements on CFRP plates (4) of size 150 mm × 150 mm (measurement area 120 mm × 120 mm) and measured in a controlled way (according to a protocol) after the BL surface treatment condition; A) CA values range; B) Q-Q plot for results from four plates. [8]

3.2 Shear strength per CA RI for non-conditioned (ambient) specimens

To correlate specimen (series) CA levels with shear strength (SLS specimens), tests in ambient conditions were performed: average maximum (over series) stress for BL was 30.5 ± 1.8 MPa, and average maximum (over series) stress for grease contaminated joints was 37.7 ± 4.0 MPa. The results show that FM 300-2 is not especially sensitive to the GR contamination and the mechanical strength results are almost comparable within standard deviations. Both failure modes were essentially cohesive in the adhesive layer. This means that SLS testing at ambient conditions cannot clearly indicate the contamination or lower surface treatment quality.

3.3 Shear strength per CA RI for weather chamber-conditioned (ETW) specimens

The results comparing the effects of the ETW conditioning on SLS results are in Figure 4; the stress values are evaluated based on individual bond surface per specimen. The conditioning resulted in clearly (more than two standard deviations) lower GR shear strength, i.e., the test method tended to show the effect of grease contamination. The average maximum (over series) stress (for nominal bond area) for BL was 27.2 ± 0.3 MPa, and average maximum (over series) stress (for nominal bond area) for grease-contaminated (GR) joints was 23.0 ± 0.1 MPa. The grease application for the GR series tends to have improved the effective ductility of the joint and, therefore, increased the peak force recorded during the test (RTD and ambient testing conditions) with respective increase in the average (lap shear) strength.

For the ETW series, i.e., after specimen conditioning, the strength values were lower for both BS and GR series compared to the RTD testing, as is expected due to the typical softening of adhesive due to moisture. However, the determined lap shear strength difference between BL and GR specimens is surprisingly small after the ETW conditioning. The fracture surfaces for all the

specimens were essentially identified as cohesive failure in adhesive or interlaminar (for GR RTD series) in CFRP; some minor zones could have been identified as adhesion failure. As summary, no observably significant difference was detected between any of these series based on the failure mode or challenge related to interfacial adhesion.

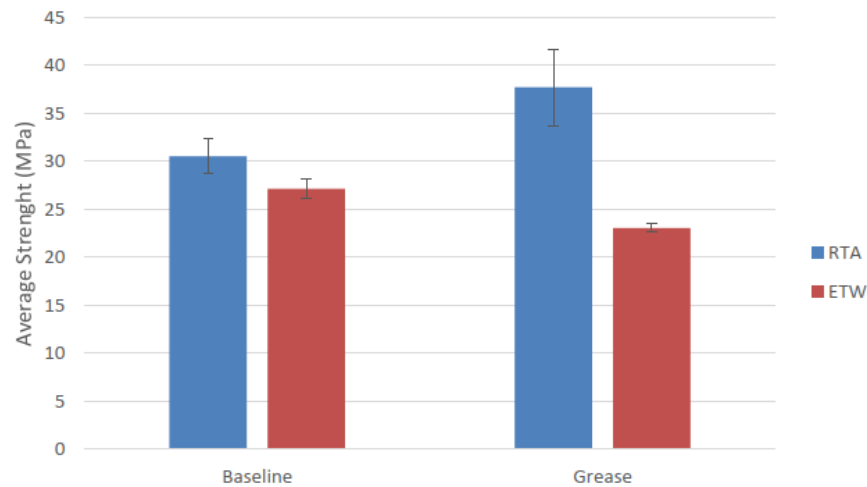


Figure 4 – SLS shear strength results for BL (left, Baseline) and GR (right, Grease) surface series when the testing was carried out for non-conditioned (RTA) specimens and ETW-conditioned specimens (ETW). [10]

3.4 Shear strength per CA RI for water immersion-conditioned (ETWi) specimens after ETW

Finally, the effect of the continued (after ETW done) water immersion (i.e. condition ETWi) was studied in terms of bond strength. The average maximum (over series) stress for BL was 26.7 MPa, and average maximum (over series) stress for grease contaminated (GR) joints was 23.6 MPa. The failure mode for both series was essentially cohesive in the adhesive layer (Figure 5). Based on the results, the continued water immersion after ETW did not significantly increase the sensitivity of the SLS method to distinguish between two completely different surface qualities when compared to the results of the series with only ETW condition. The shear strength for the BL series changed -1.9% (0.5 MPa) and the shear strength of GR series increased 2.4% (0.6 MPa) (toughening effect). The changes are above standard deviation of the stress values from the ETW series testing.

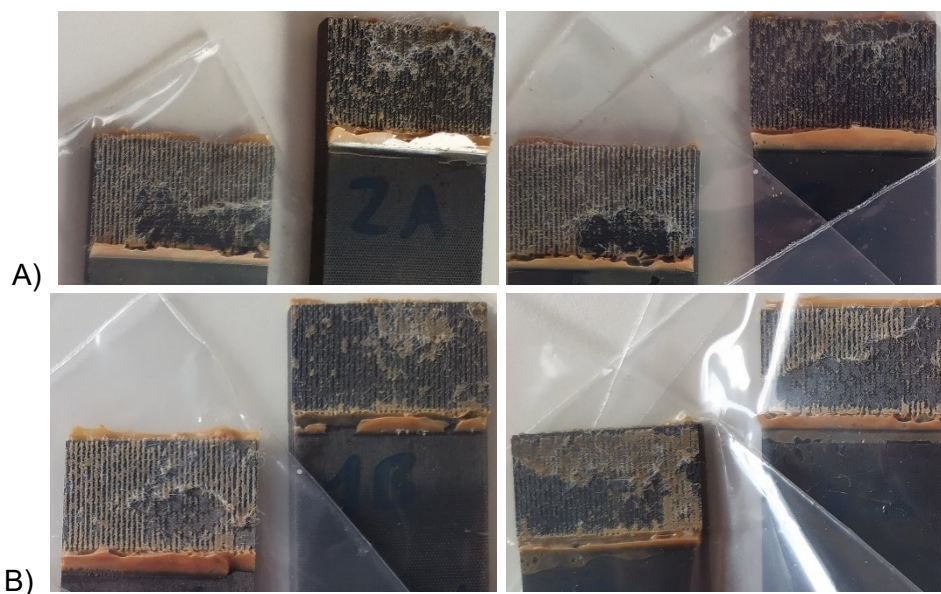


Figure 5 – Failure mode images after shear testing BL and GR series when the testing was carried out for weather chamber-conditioned and also water immersion-conditioned (ETWi) specimens; A) BL series; B) GR series. The specimens in the figure are kept in storage bags for later fracture surface characterisation. The specimen (adherent) width is 1 inch (for scale).

3.5 DCB test method for trial testing of surface conditions

The DCB testing was performed to find if fracture testing could be used as a method to correlate certain surface quality and respective CA (range) with the mechanical (bond) strength. It is well known that mode II test methods tend to result in interfacial crack paths, i.e., the crack trajectory is forced against the bi-material interface [11][12]. The DCB testing leads to essentially pure mode I fracture, but the use of the pre-crack and damage localization was here believed to be a solution for a higher sensitivity to the applied surface treatments (or contamination for the GR series). The failure surfaces of the DCB test specimens of the GR series are shown in Figure 6.

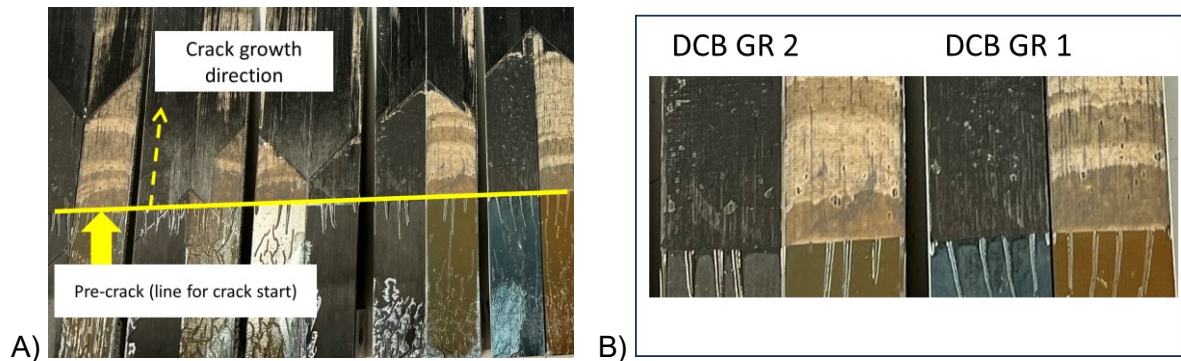


Figure 6 – Failure mode images after DCB fracture testing (RTD) of GR series joints: A) full series (adherent pairs side-by-side, fully detached); B) detail of crack growth for the first 30 mm for the example specimens DCB GR1 and DCB GR2. Specimen width 20 mm (for scale).

From Figure 6, it can be seen that the fracture mode is mainly adhesion failure. Only after the full manual detachment (post-test task for some specimens by the researchers) of adherents from each other, failure mode switched to interlaminar. Harsh removing of peel ply prior to surface treatments was deemed part of the reason for the interlaminar mode for the later crack path. The removal by sharp tool had cut some fibers that might have led to interlaminar fracture at the bond area left after actual testing.

4. Conclusions

This work focused on comparing the capability of shear lap testing (here with acronym SLS) to distinguish specific surface qualities in terms of significantly different average strength of test series. The quality indicator for each surface treatment was a contact angle measurement in this work and the distribution of CA values was reported. The SLS testing was carried out for three different statuses of specimens: 1) no conditioning (RTD), 2) elevated temperature wet (ETW) by weather chamber (15 days with elevated temperature (60 °C) and moisture (90-95% RH)), 3) water immersion at elevated temperature (60 °C) for 27 days after the ETW conditioning done. With these statuses, two different surface qualities were considered: 1) typical mechanical roughening; 2) excessive grease contamination after the typical roughening. Also, fracture testing with the DCB specimen was studied for the contaminated series to trial its use for adhesion evaluation related to certain surface quality. The results indicated the following conclusions:

- Contact angle measurements indicated significantly lowered surface quality for grease-contaminated CFRP surfaces compared to non-contaminated counterparts.
- The applied aerospace grade adhesive (FM 300-2) was not especially sensitive to grease contamination when SLS tests were performed without specimen conditioning (i.e. RTD tests); no degradation was observed in adhesion.
- After ETW conditioning, the lap shear strength of grease-contaminated specimens was clearly lower than that of non-contaminated specimens in terms of series average.

- Continued water immersion (ETWi) after the ETW conditioning tended not to have marked changes in lap shear strength values compared to the ETW series even when noting the low number of pieces tested in this condition.
- Fracture testing with DCB specimens had observably clearer switch of failure mode due to the grease contamination treatment compared to the SLS specimens yet the complexity with interlaminar failure mode will require further analysis.

Acknowledgement

The authors are grateful for the support and funding of this research by Finnish Defence Forces Logistics Command.

5. Contact Author Email Address

Mikko Kanerva, mailto: mikko.kanerva@tuni.fi

6. Copyright Statement

The authors confirm that they, and/or their company or organization, hold copyright on all of the original material included in this paper. The authors also confirm that they have obtained permission, from the copyright holder of any third party material included in this paper, to publish it as part of their paper. The authors confirm that they give permission, or have obtained permission from the copyright holder of this paper, for the publication and distribution of this paper as part of the ICAS proceedings or as individual off-prints from the proceedings.

References

- [1] Molitor, P., Barron, V., Young, T. "Surface treatment of titanium for adhesive bonding to polymer composites: a review", *Int J Adhes Adhes* 2001;21(2):129-36.
- [2] Brockmann, W., Hennemann, O.-D., Kollek, K., et al. "Adhesion in bonded aluminium joints for aircraft construction", *Int J Adh Adh* 1986;6(3):115-43.
- [3] Aakkula, J., Saarela, O., Haikola, T., et al. "DIARC plasma coating for reliable and durable structural bonding of metals", In: *Proceedings of the ICAS 28th Congress*, 2012.
- [4] Bènard, Q., Fois, M., Grisel, M. "Roughness and fibre reinforcement effect onto wettability of composite surfaces", *Appl Surf Sci* 2007;253(10):4753-8.
- [5] Chin, J., Wightman, J. "Surface characterization and adhesive bonding of toughened bismaleimide composites", *Composites Part A* 1996;27(6):419-28.
- [6] Flinn, B.D., Clark, B.K., Satterwhite, J., et al. "Influence of peel ply type on adhesive bonding of composites", In: *Proceedings of the international Sampe symposium and exhibition*, Baltimore, USA, 2007.
- [7] Bènard, Q., Fois, M., Grisel, M. "Peel ply surface treatment for composite assemblies: chemistry and morphology effects", *Composites Part A* 2005;36(11):1562-8.
- [8] Mews, F. "Identification of CFRP surface condition prior to bonding". M.Sc. thesis, In: *Trepo. Tampere University*. 2023. <https://urn.fi/URN:NBN:fi:tuni-202306226896>
- [9] Brighton Science, "Handheld Surface Analyst". <https://www.brighton-science.com/solutions/non-networked-solutions/5001-high-performance-surface-inspection> (accessed Dec 2023).
- [10] Blanco Garde, D. "Correspondence between adhesive joint strength, surface treatment and surface quality of CFRP". M.Sc. thesis, In: *Trepo. Tampere University*. 2023. <https://urn.fi/URN:NBN:fi:tuni-202306226895>
- [11] Chen, B., Dillard, D., Dillard, J., et al. "Crack path selection in adhesively bonded joints: the roles of external loads and specimen geometry", *Int J Fract* 2002;114(2):167-90.
- [12] Choupani, N. "Interfacial mixed-mode fracture characterization of adhesively bonded joints", *Int J Adhes Adhes* 2008;28(6):267-82.