



# Reliable Adhesive Bonding of Carbon Fibre Reinforced Plastics (CFRP) for Manufacturing and Repair

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#### ABSTRACT

High performance fibre reinforced composites are increasingly used in light weight aerospace structures. The high tensile strength, the low weight, the favourable fatigue behaviour, and the ruggedness against out- err influences (corrosion) have led to a significant increase in the use of carbon fibre reinforced composites (CFRP). Beside various successful application in military aviation, nowadays there are even two civil airliners (Boeing 787 and the Airbus A350) on the market that mainly consist of CFRP materials.

Adhesive bonding is a powerful joining method that engineers would like to use for structural bonding of carbon fibre reinforces plastics for manufacturing and repair. Key factor for the quality of bonded joints and Achilles heel is the phenomena adhesion. Without a realistic chance for a non-destructive testing method for the quality of the produced adhesion, the only possible way forward is a robust and therefore reliable bonding process.

To reach this challenging goal, adhesion mechanisms and influencing factors on the bond quality have to be understood in adhesion relevant dimensions. Surface nano-analysis and destructive tests with variation of surfaces are mainly used to build up the necessary knowledge.

For manufacturing and repair, strategies for reliable use of the nanotechnology adhesive bonding in industrial environment were developed.

# 1.0 GENERAL COMMENT ON STRUCTURAL ADHESIVE BONDING OF CFRP

Adhesive bonding has already been applied for many years with great success in the manufacturing and repair of fibre-reinforced structures although it is still a challenge is to establishing reliable and robust adhesive bonding processes. Moreover, there is no non-destructive technique that can quantitatively test bonded structures. And, for repair processes on damaged aircrafts with used material and the presence of numerous contaminants the situation is even worse. From the authors point of view, the key factor for successful adhesive bonding of CFRP parts is the creation of proper surfaces.

#### 1.1 Surface treatment of CFRP

To achieve a structural and ageing resistant bonding the formation of adhesion forces between adhesive and adherent have to be guaranteed. During the production process of CFRP internal and external release agents



are applied. Therefore surface free energy is reduced and a nonpolar surface is the result. Hence a surface pretreatment is necessary to clean the surface by a removal of the release agents. Furthermore the literature often mentions an activation of the surface; this means the accumulation of chemical groups at the surface. For the preparation of CFRP there exist various possible surface treatments, which can be divided in physical, mechanical and chemical processes. Chemical and physical surface treatments cause an increase of surface free energy by the attachment of polar respectively reactive chemical groups. A well-known physical surface treatment is the atmospheric pressure plasma jet, where oxygen groups are attached at the polymeric surface. Thereby a high polar surface with high strength bonds can be ensured. However with mechanical surface treatments release agents are removed from the surface and a "fresh" polymeric surface is generated.

Mechanical processes like grinding have been successfully deployed for years in the repair of CFRP structures. To qualify and optimize bonding processes surface free energy, roughness and destructive testing are performed in an iterative process.

#### 1.2 Appropriate use of destructive test to quantify adhesion

Due to the lack of non-destructive testing methods, destructive tests have to be undertaken. For CFRP, various destructive test setups exist. Double cantilever beam (DCB), floating roller peel, and single lap shear specimens are commonly used to qualify processes and to quantify adhesion strength. Despite the lack of stress concentration for the boundary layer in these destructive test methods, there is always the problem of the limited laminate strength within CFRP. The high strength adhesives exceed the strength of the CFRP fiber-matrix interface. Therefore it is difficult to test the quality of the adhesion and to compare different surface treatments.

One example is to vary the joint length of single lap shear specimens. With an decreasing joint length, the stress distribution is more uniform. Figure 1-1 shows the lap shear shrength of APPJ treated single lap shear specimens with various joint length (6,25 mm, 12,5 mm, 25 mm). A decreasing joint length results in an increase of lap shear strength from 22 MPa (25 mm) up to 50 MPa (6,25 mm). In addition, these joint length variation results in a change of the fracture pattern from a light fibre tear (6,25 mm) to a cohesive failure in the adhesive. So with a more uniform stress distribution at a joint length of 6,25 mm it is possible to generate high strength bondings with a cohesive fracture pattern. Now the shear stresses determine the failure behavior and the adhesive bond can be tested up to 50 MPa.



Figure 1-1: Lap shear strength and fracture surfaces of APPJ-treated single lap shear specimens with various joint length.

For structural applications, also the longtime durability of the adhesion has to be guaranteed. To obtain a statement about the longterm stability of a bonded joint hydrothermal aging tests are necessary. For aging tests of APPJ-treated single lap shear specimens, again the fiber-matrix adhesion is the archilles heel of the



adhesive joint. The fiber-matrix adhesion decreases while hydrothermal aging (85°C, 85 % r.H., 1000 h). This causes a laminate failure of CFRP after aging (Figure 1-2). It is therefore not possible to get any information about the quality of adhesion. The same behaviour can be observed for DCB samples.



Figure 1-2: Lap shear strength and fracture surfaces of single lap shear specimens before and after hydrothermal aging (85°C, 85 % r.H., 1000 h) [4]

To determine the stability of the adhesion generated by different surface treatments a different way of testing is proposed: To exclude the fiber matrix adhesion, by using adhesivly bonded pure matrix resin samples.

# 2.0 ADHESIVE BONDING FOR MANUFACURING

For manufacturing, bondable surfaces need to be created in a reliable way. But aircraft parts are manufactured all over world today. Structures are therefore can be handled several times, transported hundreds of kilometres, stored for months in unclear environments and examined (e.g. with ultrasound). A surface treatment prior to bonding is therefore a must. Manufacturing of the composite structures has a big influence on the initial CFRP surfaces.

#### 2.1 The use of peel plies

Peel-plies are widely used for the fabrication of CFRP structures. They have some important advantages. First, they sponge up the surplus resin during manufacturing. Afterward they cover the surface during nondestructive testing of the fabricated structures, handling and for storage. They can be removed instantaneous prior to adhesive bonding to create "fresh and bondable" surfaces.





Figure 2-1: Topography (SEM images) and surface chemistry (via XPS) of the used peel plies is transferred due to the manufacturing process from the peel plies onto the CFRP surfaces [1].

But in reality, common peel-plies are coated with release agents to avoid fibre pull out during peel-ply removal. This leads to contaminated surfaces [1], low bond strength and inhibits a reliable use of adhesive bonding for safety relevant structural applications. Figure 2-1 exemplary shows CFRP surfaces produced with two different peel plies. The CFRP specimens were made from a prepreg material (Hexcel 8552/IM7 with epoxy matrix) in an autoclave process. They were manufactured to form a quasi-isotropic layup of 2 mm in thickness. In neither case a surface suitable for a reliable adhesive bonding was created. Hence, using peel-plies is a method to create reproducible, but contaminated surfaces. Only in combination with a surface treatment process it is possible to establish reproducible surfaces. Especially plasma processes are very interesting because they allow to create a well-defined surface chemistry and topography on the nanometre scale.

Our research shows that, the combination of peel-ply and subsequent plasma treatment allows the creation of a reproducible surface on nanometre scale even in industrial processes. In order to accomplish this reliable surface treatment, extensive research activities on plasma composition and effectiveness on CFRP surfaces [2] in combination with intensive optimization of peel plies were necessary. To test the strength and the durability of the adhesive bonds, suitable destructive tests have to be performed to test especially the interface.

## 2.2 Combined use of peel plies and atmospheric plasma jets treatment

Atmospheric pressure plasma jets (APPJ) are often used to treat a surface line by line to modify the entire area intended for bonding. Using compressed air as process gas and a manipulator to move the nozzle, it is a keen technology that can be integrated into an automated process chain. Depending on the process parameters (nozzle-substrate distance, movement speed etc.) the intensity of the APPJ- treatment can be varied.

Scanning Electron Microscopy (SEM), Laser Scanning Microscopy (LSM) and Atomic Force Microscopy (AFM) are used to measure topography changes qualitatively and quantitatively. Contact Angle Measurement and X-Ray Photoelectron Spectroscopy (XPS) allow investigations on the chemical composition of the very top atomic layers of CFRP surfaces. Figure 2-2 shows how a CFRP surface (Hexcel 8552/IM7 manufactured with a fluorine-based release foil) is changed by an APPJ- treatment.





Figure 2-2: Differences between an untreated (left) and APPJ- treated (right) CFRP- surface. Changes in topography can be seen in SEM (top) and especially in AFM (bottom) images. Chemical modification can be seen in the surface free energy (middle, bottom) and XPS- results (middle, top).

The topography on the micrometre scale, created by the peel ply used, is affected very little while on the nanometre scale a much rougher surface with an increased surface area is created. The chemical modification is distinguished by the removal of release agents' residuals and the attachment of oxygen containing functional groups.

An improvement in bonding strength through APPJ- treatment can be achieved within a wide range of process parameters, avoiding interfacial failure, for different types of peel plies and release foils as well. For a variety of fluorine-based peel plies a change in failure mode from interfacial to cohesive/ laminate failure accompanied with increased bonding strength can be observed after APPJ- treatment, even if the fluorine is not totally removed [5].

Detailed investigation has been carried out to improve the understanding of the plasma-surface-interaction. Thus, not only areal treated specimen are investigated but single line treatments as well [2]. If a fluorinebased peel ply (or release foil) is used in the manufacturing process, the fluorine-carbon ratio and the oxygen-carbon-ratio determined via XPS can be used to indicate the cleaning and activation effect on the surface as shown in Figure 2-3. Dependent on the used process parameters differences in intensity and width can be shown. Comparing these results with analysis of the plasma itself allow control of the process control that is independent of the used APPJ-system and thus transferable to different systems.





Figure 2-3: Single line APPJ-treatment of a specimen (left) and distribution of the O/C-ratio (right, top) and the F/C-ratio (right, bottom) for a nozzle-substrate distance of 8 mm (blue) and 12 mm (red) [2].

## **3.0 AUTOMATED SURFACE TREATMENT FOR REPAIR**

As first step, cleaning is a must to avoid smearing and spreading of mobile contaminants on the surface. For the past decades up to now, areas for repair are prepared by hand tools using sand paper or sanding discs. It is a simple method to remove thick layers of non-bondable, undefined surface layers. But the sanding processes are time consuming and highly dependent on the personal skill-level. The creation of a scarf by hand requires several hours. Visually exact stripping even of thin layers is possible. But it has to be taken into account, that grinding of polymers often does not lead to bondable surfaces and the ability to achieve satisfactory bond strength. Sanding therefore is a problematic process that is furthermore difficult to automate.

#### 3.1 Automated Milling

In preliminary studies, different surface preparation technologies like blasting, sanding, laser and milling have been examined by surface analysis methods and destructive tests.

Milling is a process that allows material to be removed with high precision. In contrast to grinding and abrasive processes, milling is capable of reproducibly creating uniform surfaces at a high level of automation. Therefore, it has been chosen for the automated repair. In order to produce CFRP surfaces suitable for adhesive bonding, i.e. without smearing and minimal interference to the fibre-matrix interfaces, the milling process needs to be optimized. Due to the material properties of cfrp and all types of composites in general, it is challenging to find a proper milling strategy to yield suitable surfaces. Because a composite comprises at least two different materials, the cutting conditions for the tool are constantly changing. Furthermore, the deterioration of the milling tools is a challenge.





Figure 3-1: PM3 (left [6]) and FMRS (right) mounted on aircraft fuselage structures

In order to improve cost effectiveness and reliability, automated 5-axis milling tools for the repair area have been developed by the German armed forces in close co-operation with industrial partners that allow a fast and reproducible creation of scarfs with highly bondable surfaces. The first prototype PM3 was build up in 2010. The actual mobile milling machine of the WIWeB, the Future Mobile Repair System (FMRS) was delivered in 2014 (figure 3-1). The frame structure of the FMRS and several parts are made of CFRP to achieve sufficient stiffness for the milling process at low weight. By installation trials on real aircrafts, the adaptation capability was approved.

The mobile repair system consists of a mobile 5-axes-milling unit and a control station. The mobile milling unit can be mounted on the damaged aircraft structures by means of a vacuum suction system. The entire repair process can be operated from a computer-based control centre using a graphical user interface. An integrated laser-line scanner allows sampling of the complete surface with high accuracy.

After setting the process data and the type of repair (e.g. scarf geometry, layer thickness) three axes milling program is produced. This three-axes milling program is then adapted geometrically to the scanned surface and translated into machine code for the five axes kinematic (figure 3-2).



Figure 3-2: Laser scanning (left [1]) and milling of a rectangular scarf (right [6])

The FMRS is a technology demonstrator that is used in a combined research program of the WIWeB to investigate possible improvements for the repair. Additional technologies like ultrasonic non-destructive testing, a plasma torch for surface activation or a power ultrasound adhesive primer application can be adapted to the 5-axis kinematic.



#### 3.2 Automated scarfing allows new kind of repairs

The repeatable machining of laminates guarantees a defined geometry and more uniform surface quality and can also be used to realize complex and optimized repair geometries [7]. Currently used scarf repair patterns are of simple circular, rectangular or elliptical shapes with increasing dimensions towards the structure surface. With the recently developed automated milling systems, it became possible to machine ply-wise stepped repairs with individual shapes into laminates.

The idea of fiber-oriented repair geometries in aircraft composite structures aims to reduce the repair area by considering the orthotropic properties of reinforced plies. In Figure 3-3, a circular damage is used to explain the idea of fiber-oriented repair geometries.



Figure 3-3: Graphical comparison between a fibre-oriented repair geometry on the right and a circular repair geometry on the left. Both repair geometries are based on a circular damage of 10 mm in diameter in the center of the repair, a (45/0/-45/90)S lay-up and identical step lengths. The dotted outmost contour shows the top layer of the repair [7]



As a first step, tensile tests of a stepped lap joint were done [7]. For the specimens with constant step lengths, an overlap of 6.25mm was chosen for every step (scarfing ratio of 1:50). In fiber-oriented specimens, plies oriented in 0-degree direction had full overlap lengths of 6.25 mm, while plies oriented in +45 or -45 degrees had an overlap length of 4.42 mm. No overlaps were designed in plies with 90-degree fiber orientation. Carbon fiber UD reinforced epoxy prepreg Hexcel HexPly 8552/IM7 was used to manufacture quasi-isotropic laminates with a  $(0^{\circ}/45^{\circ}/90^{\circ}/-45^{\circ})$ 2S lay-up and a layer thickness of 0.125 mm. No adhesive was used to exclude effects from bonding processes.



Figure 3-4: Schematic drawing of the step lengths in the investigated specimen series (left; all values in mm). For easier understanding, only the lowest four plies of the investigated laminates are displayed; Mean values of determined tensile strengths (right) of the particular series. Borders of two standard deviations are indicated around the mean values [7].

For fibre-oriented joint specimens with and without overlaps, similar mean tensile strength was measured (figure 3-4). Reducing step lengths between UD plies with fibres not oriented in load direction, had no significant influence on the tensile strength of the stepped joints.

# 4.0 CONCLUSIONS

The use of structural adhesive bonding of CFRP is a challenge. For proper surface-adhesive interaction surfaces have to be prepared in a defined way. By using a combination of peel-plies and atmospheric pressure plasma jet this goal can be reached in an industrial environment. Therefore, the effectiveness of the peel ply and the plasma treatment can be understood on the nanometre scale. Plasma composition and peel-plies have to be optimized to establish a reliable adhesive bonding process with a maximized process window. Destructive tests are necessary and must be able to test the strength and durability of the generated adhesion.

The applicability is shown within the "FFS" (Fortschrittliche Flugzeug Strukturen) project, a close cooperation of WIWeB, DLR, and AIRBUS D&S in which an airbrake of a combat aircraft is rebuild with structural adhesive bonds and certified for military airworthiness. Automation of the CFRP repair helps to speed up the repairing process significantly and to improve the quality of bonded joints and reproducibility. Furthermore, automation may allow the realization of more efficient geometries for the repair of CFRP. By this, a reduction of repair geometries can be reached.



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