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Adhesion properties of the hybrid system made of laser-structured aluminium EN AW 6082 and CFRP by co-bonding-pressing process

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A parameter investigation for manufacturing a hybrid system through the prepreg pressing process was carried out within the scope of this work to achieve optimal adhesion properties. The hybrid specimen comprises an aluminium sheet of alloy EN AW 6082 in T6 condition and a thermoset Carbon Fibre Reinforced Plastics prepreg. The prepreg pressing process allows the curing reaction of epoxy resin and the joining process to occur simultaneously to avoid an additional bonding process step. The surface of the aluminium sheet was pretreated in advance using a pulsed Nd:YAG laser to enhance the bonding properties. In the first step, the shear edge tests investigated the adhesion properties achieved with different consolidation (temperature, time and pressure) and laser parameters. Then, 3-point bending tests were carried out to investigate the influence of the consolidation parameters on the mechanical properties of the Carbon Fibre Reinforced Plastics-laminate. In this way, the optimal parameter sets for manufacturing hybrid structures were determined.

Keywords: prepreg pressing process; laser surface pretreatment; hybrid material, adhesive bonding

1. Introduction

Vehicle mass has increased significantly in recent years due to increased demands for safety and comfort.[1] Simultaneously, requirements for climate protection in Europe have become stricter, with Greenhouse Gas emissions to be reduced by 55% by 2030.[2] Higher mass in vehicles increases energy consumption and thus the emission of greenhouse gases. Therefore, the development of innovative lightweight design approaches is of high importance. Since a reduction in vehicle mass lowers necessary drive power, lightweight construction also plays a significant role in electromobility. For example, smaller and thus lighter batteries or electric motors can be used with the same performance, which further reduces the mass and thus lowers the loading of the structures.[3]

Various lightweight design approaches have been developed to achieve climate and lightweight design goals. Multi-material design plays an essential role in exploiting the advantages of different materials, such as aluminium and Carbon Fibre-Reinforced Plastics (CFRP). This way, reduced weight and improved mechanical properties could be achieved simultaneously.[1] On the other hand, it must be mentioned that hybrid construction will also place significant demands on the manufacturing and joining process. High differences in the electrochemical potentials between Fibre-Reinforced Plastic (FRP) and aluminium and the exposure to environmental influences renders the corrosion properties of such hybrid materials challenging and require suitable corrosion protection.[4]

Importantly, for reducing the manufacturing times and cost of the hybrid components out of metal and FRP, the joining technology must also be considered an integral part of the processing chain since curing is usually a time- and energy-consuming step. Currently, various techniques can be used to join metal with fibre-reinforced plastics. It can be divided into cold or thermal joining technologies. Bolted joining, riveted joining and adhesive joining are, for example, part of the cold joining technologies.[5] However, conventional joining technologies such as bolted joints and rivets can only be used at the cost of accepting significant disadvantages such as additional weight, sealing problems, and stress concentration at the holes.[6] In contrast, adhesive bonding is a beneficial solution that avoids damaging the joining partners. Nevertheless, the adhesive joining normally requires a long curing time and weakens at higher temperatures. Machado[7] investigated the adhesion properties of CFRP/aluminium joints at different temperatures using statistical and dynamic tensile shear tests. The two materials were bonded together using crash-resistant epoxy-based adhesive. Two aluminium alloys, 5754-H22 and 6060-T6, were used. The CFRP sheet consisted of 14 layers and was produced by press molding at 130°C 1h. Both aluminium joints (alloy 5754 with alloy 6060) and aluminium/CFRP joints (alloy 5754 with CFRP, alloy 6060 with CFRP) were investigated. The results showed that the CFRP/aluminium joints have higher shear stress at failure (ca. 16MPa at 24°C), while the aluminium/aluminium joints have the lowest (ca.12MPa at 24°C). This could be due to the comparatively lower strength of aluminium, as the weakest material dominates for hybrid joints failure. Moreover, it is noticed that with the increase of test temperature, a cohesive failure is seen in the adhesive as the strength of the adhesive deteriorates with the temperature increase to 80°C. To further reduce the weight of hybrid joints, for example, in the conventional rivet joining process. Huang has developed a new joining process.[8] Hybrid joints made of CFRP prepreg and aluminium were first bonded with an epoxy adhesive. After the bonding process, the hybrid joints were embossed to realize a plastic deformation of the hybrid joint. Thereby, the pressing force and the temperature of the embossing process were investigated. In contrast to conventional joining methods, such as rivet joining with an adhesive, the proposed method does not require additional components. The optimal embossing depth and temperature were 1.8mm and 100°C. In addition, an increase in force at the failure of 89.3% could be achieved.

Apart from the cold bonding technologies, many researchers have also investigated different thermal joining processes for hybrid joints, such as laser welding, friction-based welding technologies, ultrasonic welding, induction joining, and in-situ technologies. However, many of these technologies were useable only for thermoplastics.[5] To solve this problem, Lionetto used PA6 films with different thicknesses in the aluminium (alloy 5754)/CFRP joints and fabricated them by ultrasonic welding.[9] The CFRP/epoxy laminate was manufactured by vacuum bagging 9 plies of CF/epoxy prepregs stacked, adding the PA6 film as a last ply. The curing process takes at 125°C for one hour. The dependence of the adhesion properties on welding force and welding energy was investigated by tensile shear test. The PA6 film plays an active role in promoting adhesion without damaging the carbon fibres. A film thickness of 100mm is the most appropriate to obtain hybrid joints characterised by a high tensile shear strength. In addition, the welding energy and forces strongly influence the mechanical bonding at the microscopic level. However, the disadvantage is the high temperature during welding (approx. between 350-400°C) which could lead to degradation of the epoxy resin and higher thermal residual stresses in the hybrid joints.

Furthermore, laser pretreatment is particularly suitable for cleaning metal surfaces and achieving sufficient adhesion strength because of its easy handling and high geometrical freedom, which is also used in this work. The increased adhesion can be attributed to surface enlargement and increased mechanical interlocking between metal and adhesive.[10,11] Recently, various studies have dealt with the influence of laser pretreatment on the adhesion strength of FRP/metal joints. For example, Zinn et al.[12] investigated the influence of laser pretreatment on the adhesion properties of CFRP/steel hybrid composites produced by vacuum-assisted resin transfer molding (VRTM). In this process, the shear stress at failure was almost doubled. Furthermore, Heckert et al.[13] used laser pretreatment to create macro- to nanostructures on the aluminium alloy EN AW 6082 surface. GFRP was joined subsequently to pretreated EN AW 6082 by laser-based heat conduction joining. Again, a strong increase in bond strength has been observed. In addition to laser pretreatment on metal surfaces, the surface of CFRP can also be laser pretreated. For example, Schanz pretreated the aluminium and the CFRP surfaces with different laser systems (near-infrared- and ultraviolet laser).[14] Before laser pretreatment, CFRP/epoxy laminate made of 8 prepreg layers was produced by pressing at 180°C, 7 bar for 2h. Finally, the two materials were bonded with three different adhesives and the dependence of the adhesion properties on the energy density of the laser system was investigated. It was confirmed that a lower shear stress at failure can be seen with all three adhesives when the energy density is increased. Furthermore, from other literature, it is also known that the aging resistance of pure metal materials such as aluminium, steel and metal/FRP composites can be improved by laser pretreatment.[15]

Much of the research presented above works with a 2-step process, i.e., the FRP is first produced and then joined to metal by, for example, an additional bonding or welding process. Furthermore, the curing process of FRP or adhesive is also associated with a long time, such as in[7,9,14]. The manufacturing and the joining process of such hybrid material can be combined in one step to further reduce the processing or cycle times and the energy footprint. For this purpose, the prepreg pressing process is particularly suitable for manufacturing hybrid components. The prepreg pressing process is related to the sheet metal forming process, and a high degree of automation is possible.[16]

In order to achieve the objectives mentioned above, the advantages of laser pretreatment and the prepreg pressing process, a parameter investigation is carried out in this work about the adhesion properties of the hybrid composite. This study aims to achieve an energy-efficient manufacturing process in which the joining and curing processes occur in a single step. The plate-shaped hybrid composite comprises a laser-structured aluminium sheet of alloy EN AW 6082 in T6 condition and CFRP prepreg. Prepregs are pre-impregnated continuous fibres with high viscosity matrix, characterized by their easy handling, good mechanical properties, and closely tolerable fibre-matrix ratio. Due to the reactive matrix, they often have to be stored below -18°C.[17] The carbon fibres of the prepreg used are embedded in a thermoset matrix of epoxy resin. A surface pretreatment using a pulsed Nd:YAG laser is performed on the aluminium sheet before manufacturing to improve the adhesion, corrosion and aging properties. The additional bonding process is avoided, as the epoxy resin serves directly as an adhesive, reducing the process times significantly. The present work focuses on the manufacturing process and the resulting quasistatic mechanical properties of the hybrid composite.

2. Materials and Methods

EN AW 6082 aluminium alloy sheets (in T6 condition) and CFRP prepreg SIGRAPREG®C U230-0/NF-E320/39% from SGL Carbon were used to produce the specimens. The resin used in the prepreg was epoxy. The CFRP prepreg was supplied on a roll, and both sides were provided with a release film.

2.1 Manufacturing parameters

The curing behaviour of epoxy resin can vary depending on the temperature and time. In addition, the product properties are also influenced by the degree of cure. As a rule, the curing time decreases with increased temperature. Preliminary tests at the Chair of Automotive Lightweight Design at Paderborn University have identified some promising parameter sets. The results are based on a hybrid composite of the same CFRP prepreg and a steel material, S235JR. The results show that a sufficient crosslinking reaction has occurred after 90s at a temperature of 180°C. The optimum temperature range lies between 150°C and 180°C.[18] In order to determine the optimum set of parameters for the manufacturing process for hybrid composites made of CFRP prepreg and the laser-structured aluminium sheet, the test matrix is extended within the scope of this research work. For this purpose, pressing pressures of 0.3MPa, 0.5MPa and 0.8MPa with temperatures of 150°C, 160°C and 180°C were investigated. The pressing time varies depending on the temperature. Each parameter combination was combined with 3 laser parameters (L1, L2, L3; Table 1 and Figure 1) to determine interaction. Laser pretreatment with pulsed Nd:YAG-CleanLaser-CL20 (Clean Lasersysteme GmbH, Herzogenrath, Germany) was performed at the Institute of Materials Research, German Aerospace Center in Cologne. The wavelength is 1064nm, and the spot size is 65µm. For all 3 laser parameters, the laser spots overlap in both directions according to the degree of overlap specified in Table 1. The laser traverses the complete path five times consecutively for laser parameter 1 (L1 in Table 1).

Please place Table 1 here

Please place Figure 1 here

2.2 Specimen manufacturing

The specimens were produced by the prepreg pressing method. For this purpose, 6 layers of unidirectional CFRP prepreg were cut according to the specimen size and then stacked on top of each other (all at 0°). Then this multilayer composite can be placed on the pretreated aluminium sheet. Due to the tackiness of the CFRP prepreg, easy handling is guaranteed.

Please place Figure 2 here

The geometry of the specimen plate varies depending on the test methodology used later. For the shear edge tests (see section 2.3.1), the specimen plates were manufactured in a size of 70x70 mm^2 (Figure 2) and for 3-point bending tests (see section 2.3.2) in 150x150 mm^2. A hydraulic press PS200 of the company Vogt was used to produce the specimen plate, consisting of two pressure plates.

Please place Figure 3 here

Figure 3 shows the pressing tool used to manufacture the specimen plates. For the tooling (die and punch) to be better heated and automatically closed and opened, two adapter plates were manufactured, which were then attached to the press plates of the press. The die (for the shear edge and the bending specimens) was then mounted onto the adapter plate. (Figure 3) In the next step, the heating temperature, the pressing pressure and the pressing time were set. A screw-in thermocouple from TC Direct, Germany, controls the mold temperature. Once the tool reaches the target temperature, manual insertion of the prepared CFRP-aluminium hybrid composite (Figure 2) occurs. Finally, the pressing program is initiated. The lower press plate automatically moves upward until the target pressing pressure is reached. Then the pressing process and the curing of the epoxy resin occur simultaneously. Both the consolidation temperature and pressure are regulated during the pressing process. After the set pressing time, the press plate opens again, and the specimen plates can be removed. All specimen plates were post-cured in a convection oven at 180°C for 30min after manufacturing. Finally, specimens are produced from the plates by waterjet cutting.

2.3 Test methodology

2.3.1 Shear edge test

The adhesion properties between the two components of the metal-CFRP hybrid composite with varying laser and consolidation parameters were investigated. There are different ways to determine the adhesive strength of the metal-CFRP hybrid composite, for example, the short beam test according to the DIN EN ISO 14130 standard and the tensile shear test according to the DIN EN 1465 standard. However, the two methods also have some disadvantages. While failure can occur in the short beam method due to bending stress instead of shear, specimen preparation in tensile shear testing is comparatively more complex.[19] Thus, in the context of this work, following the literature by K. A. Weidenmann[19,20], the shear edge method is used to characterize the adhesion properties. Moreover, since stress concentration could occur during the test, the measured shear stress is not the true shear strength. Thus, the adhesion properties will be described within the scope of this work using "effective shear strength" in the following. The experimental setup is shown in Figure 4.

Please place Figure 4 here

The device is installed in a universal testing machine, Criterion® 45, from the company MTS system. The specimen has a width (b in Eq. 1) of 25mm and a height (h in Eq. 1) of 12.5mm. The aluminium part (3) of the specimen is inserted into the sample holder (1) and clamped between the sample holder (1) and the upper pressing plate (7) by turning the screw (6). Several sheet holders with different depths in horizontal direction have been manufactured so that samples with varying thicknesses of the metal sheet (within the scope of this work, 2mm) can be tested. Since the height of each sample may also differ minimally, the height of the pressure plate (7) can also be adjusted by turning the screw (6). The CFRP part (2) of the specimen is not clamped but only supported by the clamping screw (4) to avoid buckling of the specimen during the test. However, this clamping screw does not apply a significant force to the sample and can also be varied horizontally by turning the screw. This way, the shear plane itself can be defined. (as shown in Figure 4b) Thus, this testing procedure offers more flexibility than other testing methods. During testing, the lower part of the device moves upwards, creating a shear load at the interface. The test speed was chosen at 5mm min^{-1}. Before testing, the dimension of each specimen was measured using a calliper. Then, the effective shear strength (τ in Eq. 1) was determined analogously to the tensile shear test using the following formula, where F represents the fracture load:

$$\tau = \frac{F}{b \times h} \qquad \text{(Eq.1)}$$

2.3.2 3-point bending test

Apart from the adhesion properties at the boundary layer, the mechanical properties of the manufactured CFRP laminate are also essential criteria for determining the best set of parameters. Therefore, the laminate properties were investigated using the 3-point bending test, performed according to the DIN EN ISO 14125 standard. The specimens with a size of 100x15mm^2 were taken from the specimen plate produced by the prepreg pressing process by waterjet cutting. The bending device is installed in the universal testing machine of MTS Criterion®45. According to the standard, the support width is 80mm. The radius of the compression fin is 5mm, while the radius of both supports is 2mm. (Figure 5).

Please place Figure 5 here

The test speed is calculated according to the formula specified in the standard to 5mm min-1. Finally, the bending strength was calculated with the following formula: b is the width, h is the thickness of the specimen, and F represents the fracture load.

$$\sigma = \frac{3F}{2bh^2} \qquad \text{(Eq.2)}$$

2.3.3 Differential scanning calorimetry

Differential scanning calorimetry (DSC) was performed on CFRP prepreg to determine the degree of cure of the specimens. According to ISO 11357-1, DSC involves two test procedures in which the caloric effects of a specimen are assessed in comparison to a reference material:

- Heat-Flux DSC
- Power differential DSC

In this study, the isothermal Heat-Flux DSC, according to ISO 11357, was used. The measuring cell in Heat-Flux DSC is an oven in which the specimen and reference are heated or cooled simultaneously according to a predetermined temperature program. The temperature of both measuring points, located on a heat-conducting metal disc, is measured continuously. Then, the heat flow change is obtained from the temperature difference. Before DSC, the CFRP prepreg was cut into pieces with a mass of around 10 mg according to the standard DIN EN ISO 11357-5. The mass was determined using a precision balance XP205 from the company Mettler-Toledo. The CFRP prepreg was then positioned in the centre of an aluminium crucible. After that, the crucible was sealed. Until the measurement, the CFRP prepreg was frozen at -18°C to avoid further curing. The DSC measuring instrument DSC 214 Polyma from Netzsch was used. During the measurement, the measuring cell is first heated to the desired temperature (heating rate 10K min-1), followed by specimen insertion. After that, the isothermal measurement can be started. The holding time is 40min. Finally, the measuring cell is cooled down again to room temperature with a cooling rate of 50K min-1. The enthalpy change at different holding times can be calculated by integrating the DSC curve. In this way, the degree of cure can also be determined.

2.3.4 Characterisation of the material properties of CFRP

In the prepreg pressing process, the CFRP prepreg is pressed together with the aluminium sheet, and thus the plastic matrix is also partially pressed out. Therefore, the fibre mass-, as well as the fibre volume fraction, differs for the respective set of parameters, affecting both the mechanical properties of the CFRP laminates and the adhesion properties. For this reason, the fibre content of the specimens was determined by measuring the mass difference of the specimens after resin extraction by wet chemical methods using sulfuric acid. The measurement was carried out according to EN 2564 procedure A. After extraction, the fibres are placed in a glass filter crucible and dried at 120°C for at least 45min. Finally, the specimens are cooled, and the fibre mass fraction (W_f in Eq. 3) can be determined using Eq. 3, where m_3 is the total mass of the glass filter crucible and test specimen after resin extraction, m_2 is the mass of the glass filter crucible, and m_1 is the initial mass of the specimen.

$$W_f = 100 \times \frac{(m_3 - m_2)}{m_1} \qquad \text{(Eq.3)}$$

$$V_f = \frac{W_f \times \rho_r}{W_f \times \rho_r + (1 - W_f) \times \rho_f} \qquad \text{(Eq.4)}$$

Subsequently, the material density can then be used to convert the fibre volume fraction from the fibre mass fraction using Eq. 4. Here, V_f represents the fibre volume fraction, W_f denotes the fibre mass fraction, ρ_f and ρ_r refer to the fibre and matrix density values, respectively. In addition, the density of the cured CFRP laminate was also specified according to the DIN EN ISO 1183 standard to evaluate the state of consolidation.

3. Results and discussion

3.1 Results of shear edge test

In order to investigate the adhesion properties of hybrid samples made of laser-pretreated aluminium and CFRP, shear edge tests were first conducted. The test plan of the shear edge test is shown in Table 2. The parameter selection was based on preliminary experiments at the chair (as mentioned in section 2.1). In order to make the process more efficient, the production time in the pressing process should be as short as possible while ensuring that the samples are sufficiently pre-cured. Post-curing in the oven (180°C, 30min within this work) ensures that the samples are fully cured. Generally, a sufficient curing state could be achieved at a higher temperature with a shorter process time, as the chemical reaction is faster. Therefore, temperatures with different pressing times (150°C, 5min, 180°C, 2min) were selected in this work. In addition, a parameter set of 160°C, 20min was also selected, which was expected to be 100% cured according to the datasheet. In this way, how the pre-curing degree after the pressing process affects the adhesion properties at the respective laser parameter can be investigated. In addition to the pressing time and temperature, various pressing pressures (0.3MPa, 0.5MPa and 0.8MPa) were also investigated to examine their influence on the adhesion properties. Finally, a total of 27 parameter combinations were obtained. Five test specimens were tested for each combination. In the following sections, the specimen designation is arranged in this order: Temperature, Pressure, Time, and Laser parameters. For example, 180_0.3_2_L1 represents a specimen cured at 180°C and a pressure of 0.3MPa for 2 minutes and structured with laser parameter set L1. Preliminary tests were also carried out with aluminium sheets without laser pretreatment, in which the aluminium sheets were only degreased with acetone. However, the effective shear strength of these test specimens is very low and is around 2 - 5 MPa.

Please place Table 2 here

The results of the shear edge tests are shown in Figure 6. From the data, it is difficult to make a statement about the dependence of the effective shear strength on the laser parameter and the curing parameter. A strong dependence of the effective shear strength on pressure and temperature could only be seen for L3. No clear tendency can be found for other laser parameters.

Please place Figure 6 here

The results were then analysed using Analysis of Variance (ANOVA). The ANOVA analysis indicates whether there are statistically significant differences between the mean values of the individual groups tested.[21] The SPSS Statistics software from IBM was used to analyze the results. The measurement data were checked for normal distribution and variance homogeneity as a prerequisite. In the first step, the influences of pressure and laser parameters were considered. An interaction effect diagram is shown in Figure 7.

Please place Figure 7 here

The data points in Figure 7 show the group mean values at the respective laser parameter and pressing pressure used for specimen preparation. The error bars represent the 95% confidence interval, which indicates that the true mean values of a population are within the range of 95%. For statistical evaluation, the significance level was calculated, the limit of which is set at 0.05 in most cases. If the calculated value is less than 0.05, significant differences between the two groups can be found.[21]

The effective shear strength between the individual pressures was compared for each laser parameter, and their significance values were calculated. The value is greater than 0.05 for both L1 and L2 while less than 0.001 for L3, which means that the effective shear strength strongly depends on the pressure when the aluminium surface is pretreated with L3. These results can also be seen in Figure 6 and Figure 7. For L3, the midpoints at 0.3MPa and 0.8MPa are significantly lower than those at 0.5MPa. For all other parameters, no significant differences can be found between the pressures and the laser parameters because the error bars overlap. Thus, from the analysis, it can be concluded that the effective shear strength is lowest for L3 pretreated surfaces when the pressing pressure is 0.8MPa.

In the next step, the effect of the pressing temperature on the effective shear strength was examined (Figure 8). For L1 and L2, there is no significant difference between the group means at different temperatures because the error bars overlap. However, the low mean effective shear strength of the L3 pretreated specimen at elevated temperatures, considered for all pressures and variations of curing parameters, presents another trend.

Please place Figure 8 here

The lower effective shear strength of L3 at increased temperature and pressure is mainly attributed to the effective shear strength of the specimens consolidated with the parameter set 180_0,8_2 (180°C, 0,8MPa, 2min, Figure 6), which have the lowest effective shear strength of all specimens. The height measurements of the microstructures with the Scanning Electron Microscope (SEM) (Figure 9) indicate that the laser parameter L3 seems to generate the lowest microstructures. The resin probably cannot be absorbed by the structuring at high pressure. In addition, the resin viscosity is also lower at elevated temperatures and can thus flow better.[22,23] That could result in an increased squeeze out of the thermoset matrix in the boundary area during consolidation, leading to a higher fibre matrix ratio. A loss of the thermoset matrix is revealed by the characterization of the fibre volume fraction (using the wet chemical method according to EN 2564). At 0.3MPa pressing pressure, the fibre volume fraction is lowest at 43% and rises at 0.5MPa to 47% and 0.8MPa to 67%, indicating that less thermoset matrix remains available for the bond line. Thus, the layer thickness of the epoxy resin on the boundary layer is low, which can lead to poor adhesion.

Please place Figure 9 here

Some studies have researched the dependence of bond strength on adhesive layer thickness. For example, Yang et al.[24] investigated aluminium samples bonded with two types of adhesives: epoxy and silicone rubber, with the smallest investigated adhesive layer thickness of 0.02mm and the largest of 1mm. The results showed that the bond strength deteriorated with increasing adhesive layer thickness up to a limit of 0.4mm (with an epoxy adhesive.

However, in this work, no additional adhesive was used. In contrast to the above publications, the epoxy resin system directly serves as the adhesive. Furthermore, compared to the investigated adhesive thickness in the publications above, the thickness of the epoxy resin layer at the interface in this work is minimal (Figure 10), so the effect mentioned above does not yet occur. Thus, for the investigation with the scope of this work, a sufficient amount of resin must be at the boundary layer to ensure a sufficient bonding between the metal and CFRP. Nevertheless, the resulting effective shear strength of specimens pretreated with L1, which generates the highest surface structures, should be the highest. This is not the case for most of the consolidated specimens in this study.

Figure 10 shows the microscopy sections of the interface region of EN AW 6082-CFRP specimen (consolidation parameter: 150_0.3_5) after the surface pretreatment with the laser parameters L1 to L3. From this, it can be seen that at L1, both the epoxy resin and the carbon fibres penetrate more into the surface structuring than at the other two laser parameters. This µm-sized waviness at the interface could lead to the notch effect and stress increase in the CFRP prepreg, which tends to fail at this point. That could be why the effective shear strength at L1 is not always the highest, although the surface structuring is highest at L1.

Please place Figure 10 here

3.2 Fracture surface analysis of the shear edge specimens

The fracture surfaces are shown in Figure 11 based on the 150_0.3_5 specimen. All other specimens show similar fracture surfaces. It can be seen from Figure 11 that a completely cohesive fracture of the CFRP prepreg occurs for the L1 pretreated specimen since there is still a significant amount of residual CFRP on the aluminium surface. While the residual of CFRP-prepreg on the aluminium surface pretreated with L2 and L3 is significantly lower. Upon comparing Figure 9 with Figure 10, the same conclusion as in the previous section can be drawn. The µm-sized waviness in the CFRP laminate could lead to stress concentration, resulting in earlier failure than the connection at the boundary layer. It is possible that the adhesive strength of the samples produced with L1 already exceeds the interlaminar shear strength of the CFRP laminate, which is why the CFRP laminates fail first. Therefore, further investigations need to be conducted to determine the interlaminar shear strength of pure CFRP laminate at the respective process parameter.

Please place Figure 11 here

The fracture surfaces were then analysed by SEM. The red circle in Figure 11 shows the position where the SEM images were taken. From the SEM images (Figure 12), it can be seen that on the fracture surface of the specimen pretreated with L2 and L3, besides the fractured CFRP prepreg, there are some bright spots (in red circle). These spots could be either the metal surface or due to a stronger charge of the epoxy layer during the SEM analysis. Therefore, Energy-dispersive X-ray spectroscopy analysis (EDX) was performed on these locations to determine the material composition. Here, point 1 and point 4 in Figure 13 c) and d) are exactly the bright spots in Figure 13 (a) and (b). Points 2 and 3 are the area next to them.

Please place Figure 12 here

Please place Figure 13 here

Figure 14 shows the EDX spectrum of the single point in Figure 13 (c) and (d). It can be seen that there is still an epoxy layer on the fractured surface at points 2 and 3 since carbon represents an important material element of the epoxy resin, and the nitrogen could come from the amino groups of the hardener system. However, in opposition to that, an extreme aluminium peak can be detected at points 1 and 4. Therefore, these bright spots may be the micro- or nanostructure of the laser pretreatment, and adhesion fracture could occur at these locations.

Please place Figure 14 here

Usually, cohesive fracture of the thermoset matrix occurs if the surface pretreatment is suitable to allow enhanced adhesive bonding between the two joining parts. In this case, the maximum transmissible force is achieved.[25] Correlating the fracture surfaces with the surface texturing (Figure 9 and Figure 10), it can be concluded that the effect of mechanical interlocking on the micro-scale could be greatest for L1 pretreated specimens. However, the bond strength of hybrid composites is not only dependent on mechanical adhesion. From the previous shear edge results, it can be determined that the layer thickness of the epoxy resin, or the stress distribution at the interface, also influences it. In addition, other factors also influence the adhesion properties, such as the presence and degree of nanostructures, which could affect the wetting and infiltration of the thermoset matrix. The surface enlargement on the nano-scale and microstructure (such as crater depth) could jointly influence the adhesion properties of hybrid samples and compensate for each other, as derived in the work of Freund et al.[26] Thus, the relatively lower height of the microstructure in L2 could be compensated by a larger surface enlargement on the nano-scale, resulting in the adhesion strength of the hybrid samples pretreated with laser parameter 1 and 2 being at a similar level. On the other hand, it must be mentioned that the values of the effective shear strength of L1 could also be influenced by the interlaminar failure of the CFRP laminate (as mentioned before). Better adhesion could be achieved with laser parameter 1 if the CFRP had a higher interlaminar shear strength, which should be investigated in future work.

Moreover, the inherent cohesive strength of the adhesive, here the thermoset resin, also significantly influences the bonding properties. If the inherent strength of the polymer matrix is insufficient, the load can also not be transferred effectively from the metal component to the fibres. When the surface pretreatment has optimally improved the adhesive joint, the interfacial strength of the adhesive layer is the decisive factor for the overall bond strength. Furthermore, the stiffness of the joint components also affects the adhesion properties.[27] Therefore, the mechanical properties of the CFRP laminate were also investigated by a 3-point bending test. (section 3.4)

3.3 Degree of cure

The degree of cure of the resin system can be determined from the datasheet, but factors such as the storage time of the prepreg can affect the degree of cure. Therefore, prior to the 3-point bending test, DSC analysis was performed at the three temperatures (150°C, 160°C, and 180°C) to precisely determine the degree of cure as this can have an impact on the quality of the laminate. The temperature programs of the isothermal measurement (Heating_150°C, Heating_160°C, Heating_180°C) and the DSC curves (DSC_150°C, DSC_160°C, DSC_180°C) of individual temperature are shown in Figure 15. The same holding time, as well as the heating rate, have been used for all measurements.

Please place Figure 15 here

The strong endothermic process at the beginning of each measurement is due to the rapid heating of the specimen. Subsequently, all the curves return to the exothermic range, and the curing reaction starts after a short time at all temperatures, with the reaction starting earliest at 180°C. The curing reaction is almost complete after about 10min since no further heat is released.

Although the DSC curve exhibits a distinct trajectory at each respective temperature, the total reaction enthalpy at all three temperatures after 20 minutes is nearly identical. At 150°C, it is approximately 128 J/g, whereas at 160°C and 180°C, it is around 121 J/g. The slight difference may be attributed to the prepreg being partially cured prior to testing. Generally, the prepreg is stored in a refrigerator at -18°C. However, the resin and hardener system can react at room temperature during delivery or test preparation. On the other hand, complete curing is generally impossible since an exact stoichiometric ratio between resin and hardener can never be practically achieved. Furthermore, the mobility of the molecular chains decreases as the degree of cure increases, so complete reformation is no longer possible, as described in[28,29]. Therefore, the following "100%" curing degree represents only a nearly complete curing reaction. The curing degree-time diagram at the selected temperature is shown in Figure 16. The DSC analysis indicates that the CFRP prepreg is 100% cured after 20min at all three temperatures, while at 180°C after 2min, only about 66% curing can be achieved, and at 150°C after 5min, 78%.

Please place Figure 16 here

3.4 Results 3-point bending test

The test plan of the 3-point bending test is presented in Table 3. For each test series, five specimens were tested. Moreover, the curing level after the pressing process of each parameter set is also indicated in Table 3.

Please place Table 3 here

The results of the shear edge tests indicate that 0.8MPa pressure and 180°C temperature could lead to poorer adhesion at L3. Furthermore, thermal distortion in the hybrid composite is also greater at higher temperature. Thus, these two parameters are no longer considered. Instead, two more parameter combinations were examined, namely 150_0.3_20 (150°C, 0.3MPa, 20min) and 150_0.5_20 (150°C, 0.5MPa, 20min). According to the DSC measurement, the degree of cure here is also 100%, the same as at 160°C, 20min. The aim is to lower the temperature and thus reduce the thermal residual stresses in the hybrid composite. In addition, it can be determined whether the laminate properties still depend on the temperature, although the degree of cure is the same.

Please place Figure 17 here

Figure 17 shows the results of the 3-point bending tests. Compared to the shear edge test, there is a clear tendency for the bending strength to be higher at 0.5MPa than at 0.3MPa for all curing parameters. In order to find out whether these differences between the different parameters (here, degree of cure and pressure) are statistically significant, the bending test results were also evaluated with ANOVA analysis, and their effect diagrams are shown in Figure 18 a) and b).

Please place Figure 18 here

The ANOVA analysis (Figure 18a) underlines that a significantly higher bending strength is obtained for specimens manufactured at 0.5MPa pressure than 0.3MPa (significance value of 0.004). The density measurement also indicated that the sample density for the parameter combination 160_0.3_20 was 1.5g/cm^3, while that for 160_0.5_20 was 1.53g/cm^3. It can be inferred from this observation that a better consolidation state can be achieved with higher pressure. Because air and volatiles dissolved and trapped in the matrix could be removed better at higher pressure.[30,31] Apart from the pressure factor, the degree of cure was investigated (Figure 18b). It can be determined that the bending strength is strongly dependent on the degree of cure after the pressing process: at 78% curing, the group mean value of the bending strength is only 1122 MPa, while for 100% at 150°C consolidation temperature, it is 1223 MPa and 100% at 160°C it is 1257 MPa. By having as complete a curing rim as possible, all the reactive groups of the resin and hardener can form crosslinking points. Therefore, better stiffness and strength values can also be achieved.[32] Furthermore, the bending strength does not differ significantly at a 100% curing level, although the temperature is different.

3.5 Discussion and comparison with other studies

In this study, hybrid specimens out of a laser-pretreated aluminium sheet of alloy EN AW 6082 in T6 condition and CFRP prepreg were produced using the prepreg pressing process, and their adhesive properties were examined using the novel shear edge test. In summary, it can be determined that the adhesive properties of such hybrid specimens can be significantly improved by laser pretreatment. Zinn et al.[33] also examined the adhesive properties of hybrid specimens out of a laser-pretreated aluminium sheet (EN AW 6082) and CFRP using VRTM. The best effective shear strength of 31MPa was achieved, which was also tested with the shear edge method. Although the effective shear strength is higher than the value in this study, the manufacturing time of VRTM is relatively longer. The process takes a total of 45 minutes. In addition, a preheating of the epoxy resin system was required, which makes the process inefficient compared to the prepreg pressing process.

Additionally, it should be mentioned that the test specimens have different layer configurations. While the test specimens in this study have a unidirectional layer structure and the force introduction direction is vertical to the fibre direction (Figure 4b), the CFRP laminate in the study of Zinn et al.[33] has a layer configuration of 0°/90°/0°/90°/0°/90°. Therefore, the force introduction direction parallels the fibre direction in the first CFRP layer, which could also affect the failure behaviour. The results from Zinn et al. also showed that cohesive failure between the 0° and 90° layer mainly occurs in the laser-pretreated samples, which could be attributed to the lower stiffness of the 90° layer. The same applies to the results obtained in this study. Due to the comparatively less loadable 90° fibre direction (oriented perpendicular to the direction of force) at the interface, the samples with laser parameter 1 failed cohesively, which was not the case for L2 and L3, although the manufacturing parameters and the layer structure were the same. It can be concluded that the adhesive properties of L1 probably exceed the interlaminar shear strength of the pure CFRP laminate, which should be further investigated in the future, e.g., by applying a shear edge test to pure CFRP laminates.

In addition, Lohr et al. also investigated the adhesion properties of hybrid joints using the shear edge test. It is a sandwich structure with steel face layers and a core of polylactide. The steel sheets were both laser-pretreated and nano-coated to improve adhesion. The sandwich structure was manufactured using the injection molding process. The results of the shear edge test showed that the effective shear strength of the nano-coated samples is generally little affected by the process parameters, such as the steel sheet temperature at injection. The maximum effective shear strength was reached at about 20MPa. In contrast, the effective shear strength of laser-pretreated samples was higher (25MPa) but showed more dependence on the process parameter.[34] In the context of this work, the laser parameters also partly show a strong dependence on the manufacturing process. This is especially true for L3, which is sensitive to temperature and pressure compared to the other parameters. The effective shear strength at 180 °C and 0.8 MPa was only about 6 MPa, while the other parameter combinations show a much higher value. Furthermore, a maximum effective shear strength of 20.81 MPa was achieved in this work. (180°C, 0.3 MPa, 2min with laser parameter 2), which is comparable to the results of other research.

Moreover, to enhance the corrosive properties of hybrid joints, Stoll et al.[35] have used an elastomer interlayer in a Fibre-Metal-Laminate (FML), which has CFRP as face layers and an aluminium sheet of alloy 2024 as a core. The samples produced with an elastomer layer show almost the same effective shear strength after 96h storage in the salt spray chamber (approx. 8-10MPa). In contrast, the adhesion properties of the reference samples (FML without elastomer interlayer) deteriorated significantly. However, the initial effective shear strength of the interlayered samples (approx. 8MPa) is lower than the FML samples (close to 20MPa). Thus, laser pretreatment could also be used as an alternative method to achieve good adhesion both in the initial state and after aging. The results from preliminary tests with aluminium/epoxy joints with the same laser parameter and materials have indicated that the samples pretreated with the selected laser parameters (laser parameters 1-3) show a significantly lower decrease in the effective shear strength (2-4%) after ageing.[26] Therefore, in the future, further corrosion tests could be carried out on the CFRP/Al samples to further investigate the influence of laser pretreatment on the corrosion properties of hybrid joints.

Furthermore, Li et al.[36] investigated the adhesion properties of hybrid samples made of aluminium and CFRP. In doing so, the surface of CFRP was pretreated using an infrared fibre laser, and various groove parameters (depth, spacing) were examined. Instead of using the epoxy resin in the CFRP directly as an adhesive (as in this study), a two-component epoxy structural adhesive was used. The samples were cured in an oven at 80°C for 90 minutes and then at room temperature for 7 days to cure the adhesive. The effective shear strength was then tested using the tensile shear test. Both the depth and spacing affected the effective shear strength, with a maximum of about 11.6MPa achieved.

In comparison, the hybrid samples produced in this study with a more efficient manufacturing process achieved significantly better adhesion properties. However, it must be noted that the test method in Li et al.[36] was the tensile shear test. A lot of literature has compared, for example, the tensile shear-, short beam- and the shear edge test. For the tensile shear test, bending moments and peel stress will appear, and the stress concentration at the free edge is very high.[37]. For example, Zinn et al. have investigated the adhesion properties of hybrid specimens made of steel and CFRP prepreg with different test methods. The effective shear strength determined by the tensile shear method[12] (about 20MPa) is significantly lower than the results of the shear edge method[33] (about 45MPa), even though the manufacturing process, materials, interface layer, and laser parameters were the same. Moreover, the short beam test could only deliver the apparent shear stress, and the failure behaviour strongly influences the results. In addition, large stress concentration is also found at load nose and support. Compared to the tensile shear- and short beam test, the stress distribution in the shear edge test is more uniform, and a nearly 100% shear load is created in the boundary layer.[38,39] That is also why this method was chosen in this work. But it should be mentioned that even though the tensile/peel stresses are much smaller than the tensile shear test, small stress concentration can still be found on the free edge. These stress concentrations could lead to an earlier failure of the CFRP laminate, which is the situation of the samples produced with laser parameters 1 and 2. Thus, it could be concluded that the adhesion properties of L1 and L2 manufactured samples could be better than that of L3 so that the effect of stress concentration and the peel stress at the free edge is stronger, which leads to the cohesive failure of the CFRP laminate. Moreover, the defects in the CFRP laminate, such as pores, could also lead to a stress concentration during the test.

Apart from the test method, the stress concentration at the free end is also stronger with a thicker adhesive layer.[40] However, for the hybrid joint in this study, no other adhesive film was used; the epoxy resin of the CFRP acts directly as an adhesive, and the thickness of the adhesive layer is very low, so the stress concentration at the free edge of the specimen will not be intense. Thus, it can be concluded that the shear edge method used in this study can provide reliable results for determining the shear behaviour of hybrid specimens.

4. Summary and outlook

In this study, a parameter investigation was carried out to manufacture a hybrid composite intrinsically by prepreg pressing and prior laser surface treatment of the metal. The hybrid composite consists of an aluminium sheet (EN AW 6082 T6) and CFRP prepreg with epoxy resin as a matrix system. Furthermore, the shear edge method and 3-point bending test were used to characterize the adhesion and mechanical properties. Finally, the results were evaluated by the ANOVA method. The main findings can be summarized as follows:

- The most energy-efficient processing route within the scope of this study that leads to optimal properties of the hybrid material was found to occur at a temperature of 150°C and a pressure of 0.5MPa in a 20min cycle, achieving a curing degree of 100%. These manufacturing conditions provided a compromise in terms of the mechanical strength of the CFRP, adhesive strength of the hybrid material, thermal residual stress, and energy- and cost efficiency. Further investigations with other process times, such as 10 or 15 minutes, can be conducted to make the process more efficient.
- The effective shear strength of the specimens pretreated with L3 is significantly worse at elevated temperature and pressure. The reason for this could be the lower height of the micro-structures on the aluminium surfaces. The adhesive strength of L1 is not significantly higher than that of L2, although the microstructure of L1 is significantly higher. This could be due to the fact that the nanostructure and surface enlargement on the nano-scale in L2 is greater than that in L1 so that relatively lower mechanical adhesion in L2 can be compensated for accordingly.
- The pressure increase results in a lower fibre volume fraction: at 0.8MPa, it is 67%, 47% at 0.5MPa and 42% at 0.3MPa, indicating a strong resin loss at higher

pressure. Within the scope of this work, the epoxy resin serves directly as an adhesive in the prepreg pressing process. Thus, if the amount of resin is low, the bonding effect is influenced adversely.

- The results from 3-point bending tests show that the bending strength was higher at 0.5MPa than at 0.3MPa, both for specimens manufactured with curing degrees of 78% and 100%. The density measurement also showed that the specimen density at 0.5MPa (1,53g/cm^3) is greater than at 0.3MPa (1,5g/cm^3). Therefore, a better consolidation state of the CFRP laminate and better removal of air and volatiles dissolved in the matrix can be achieved at higher pressure.

While these studies underline the feasibility of manufacturing aluminium-CFRP hybrid materials in a single forming and consolidation step, further investigations of the bonding properties under different mechanical and environmental loads will be needed. In particular, galvanic corrosion properties, as well as fatigue behaviour, will need to be studied. Furthermore, hybrid components with complex geometry can also be manufactured with prepreg pressing process. How this forming process then affects the joint properties and surface structuring remains an open question, and further studies will be necessary to answer this question.

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Disclosure statement

The authors report that there are no competing interests to declare.

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Tables

Table.1: Parameter of laser pretreatment

Parameter	Frequency [kHz]	Power [W]	Overlap Laserspots [%]	Number of crossing
L1	60	20	10	5
L2	40	20	50	1
L3	60	15	50	1

Table 2: Experimental plan of shear edge tests

Test series	Laser parameters	Temperature [°C]	Pressure [MPa]	Time [min]
1	L1/L2/L3	180	0.3	2
2	L1/L2/L3	180	0.5	2
3	L1/L2/L3	180	0.8	2
4	L1/L2/L3	150	0.3	5
5	L1/L2/L3	150	0.5	5
6	L1/L2/L3	150	0.8	5
7	L1/L2/L3	160	0.3	20
8	L1/L2/L3	160	0.5	20
9	L1/L2/L3	160	0.8	20

Table.3: Experimental plan of 3-point bending tests

Test series	Temperature [°C]	Pressure [MPa]	Time [min]	Degree of cure [%]
1	150	0.3	5	78
2	150	0.5	5	78
3	150	0.3	20	100
4	150	0.5	20	100
5	160	0.3	20	100
6	160	0.5	20	100

Figure captions

Figure 1: Scanning electron microscope (SEM) recordings of the surface structure of laser parameters, a) L1, b) L2 and c) L3

Figure 2: Geometry of the specimen plate for shear edge test

Figure 3: Pressing tool used for the manufacturing of the specimen plate. 1: Adapter plate, 2: Die, 3: Sealing frame, 4: Punch, 5: M8 screw, 6: Thermocouple

Figure 4: a) Testing device for shear edge test. 1: specimen holder aluminium side, 2: CFRP part, 3: aluminium part (2mm), 4: clamping screw CFRP part, 5: shear edge (shown in transparent), 6: clamping screw aluminium part, 7: upper pressing plate (shown in transparent); b) shows the force introduction direction

Figure.5: 3-point bending test setup, 1: Pressure pin, 2: Support, 3: Specimen

Figure. 6: Results of shear edge test. The red error bars describe deviations between each of the specimens of a set of the test series

Figure.7: Interaction diagram of pressure and laser parameters

Figure. 8: Interaction diagram of temperature and laser parameters

Figure.9: Cross-section SEM images of wetting and bonding of the resin to the aluminium surfaces after structuring with laser parameters (a) L1, (b) L2 and (c) L3

Figure.10: Illustration of the boundary layer connection of specimen 150_0.3_5 as well as the layer thickness at L1 by means of light microscopy, surface pretreated with (a) L1, (b) L2 and (c) L3

Figure 11: Fracture surfaces after shear edge testing of specimen 150_0.3_5, surface pretreated with (a) L1, (b) L2 and (c) L3

Figure.12: SEM images of the fracture surface after shear edge testing of specimen 150_0.3_5, surface pretreated with (a) L1, (b) L2 and (c) L3

Figure.13: Measuring points of the EDX analysis for the 150_0.3_5 specimen pretreated with L2 (a), (c) and L3 (b), (d).

Figure 14: EDX spectrum of the measuring points

Figure.15: Heat flux-time curve of DSC-analysis of CFRP prepreg with different temperature programs

Figure.16: Degree of cure at different temperatures

Figure.17: Results of 3-point bending test

Figure.18: Interaction diagram of 3-point bending test, a) factor pressure, b) factor degree of cure