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A comparison of IR- and UV-laser pretreatment to increase the bonding strength of adhesively joined Aluminum/CFRP components

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Abstract

For this study two commercial, thermoset CFRP laminates (called "A" and "B") were pre-treated using IR- and UV-laser prior to adhesive bonding. The achieved surface conditions were characterized by optical methods and correlated with mechanical properties of adhesively bonded hybrid Al/CFRP single-lap joints. Two opposing effects could be detected on both CFRP laminates after IR-laser pre-treatment: strength increasing surface activation and reducing weakening of the fiber-matrix interface in near surface areas. By application of UV-radiation it was possible to activate the surface damage-free for A whereas B exhibited thermal induced damaging of the fiber-matrix adhesion comparable to IR laser treatment. Furthermore, it was shown that surface activation by laser pre-treatment strongly depends on the used CFRP laminate. IR- and UV-laser pre-treatment of A leads to a significant increase of shear strength, whereas for B even a slight reduction was observed compared to chemical cleaning with acetone which was the reference process.

Keywords: carbon fibers; adhesion; surface treatments; hybrid;

1. Introduction

Multi material design is considered the key to lightweight construction, because it chooses the kind of material for each single element of the applications that fulfils the requirements of the minimal weight. To

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meet the requirements new joining techniques have to be developed (Marsh 2014). Thereby adhesive joining has great advantages over mechanical fastening. However, adhesive joints are very sensitive to surrounding influences during the joining process (Hart-Smith 1996), especially true for thermoset CFRP as well as aluminium and its alloys. There are different approaches to clean, structure and activate surfaces prior to bonding via mechanical (Kanerva et al. 2013) as well as plasma treatment (Encinas et al. 2014; Kim et al. 2003; Li et al. 2009) With laser pretreatment cleaning, structuring and activating can be combined in one single process step of contactless processing. (Völkermeyer et al. 2011, Genna et al. 2016) However, for laser pretreatment the different interactions between the epoxy matrix and the laser radiation have to be considered. For instance, by applying IR-laser most of the radiation is absorbed by the fiber. (Leone et al. 2013; Nattapat et al. 2015; Pagano et al. 2015; Semak et al. 1997, Genna et al. 2016, Reitz et al. 2017) Investigations on UV-lasers showed, that due to its higher absorption coefficient in the epoxy matrix, the removal of the matrix is more controllable (Bénard et al. 2006; Fischer et al. 2013; Palmieri et al. 2016; Rotel et al. 2000).

Firstly, the aim of this study was to explore possible correlations between the different surface conditions caused by IR or UV laser radiation and the shear strength of hybrid SLJ specimen. Secondly, special attention was set to identify the influence of the heat affected zone (HAZ) on the fiber-matrix adhesion and on the other hand focus was set to the IR-laser-system because of the significant lower investment costs which makes them very attractive from an industrial point of view (Die Lasertechnik von Morgen 2016).

2. Experimental procedure

2.1. Sample preparation

The investigated aluminum alloy is AlMg3 (EN AW-5754). Two different manufactured CFRPs are used. The first one, named CFRP A in this work, made from a prepreg of Hexcel Hexply® M21 and 30k roving Toray fiber T800S. The second one, CFRP B, is made from Dow Voraforce 5300 and 50k roving of Zoltek fiber PX35.

For laser treatment on the one hand a IR laser with the wavelength of 1064 nm was used and on the other hand an UV laser with 355 nm wavelength. For specific laser parameters see Table 1.

Both materials were joined immediately after surface treatment. As adhesive a two-component adhesive (EP-60 by HP-Textiles) was used. (Reitz et al. 2017)

Table 1. UV- and IR-laser parameters for surface preparation of CFRP and AlMg3.

Material		**CFRP**					**AlMg3**
Characteristics	Unit	UV1	UV2	IR1	IR2	IR3	IR18
Wavelength	nm	355	355	1064	1064	Treatment with once IR1 and subsequently three times IR2	1064
Average power	W	4.9	4.9	20	12		20
Pulse duration	ns	10	10	200	30		100
Frequency	kHz	20	20	27	100		45
Scan speed	mm/s	250	500	270	5000		117
Spot size	µm	35	35	104	104		67

2.2. Microscopic imaging and mechanical testing

To analyze the samples and to correlate the surface properties of the CFRPs with the mechanical properties, like shear strength, different tests were performed, both optical, contactless as well as mechanical tests according to current provisions of DIN. For more details see Reitz et al. 2017.

3. Results

3.1. Microscopic characterization of pretreated CFRP surfaces and fiber-matrix adhesion

The reference surfaces – cleaned and activated with acetone – of CFRP A and B are displayed in Fig. 1 (a) and (g). In both materials no fibers could be detected prior to the laser pretreatment process. However, laser treatment with different sets of parameter causes different surface structures for both CFRPs. With the high-energy parameter sets IR1, IR3 and UV1 a surface can be generated for both CFRPs, where the fibers are exposed as showed in Fig. 1 (b), (d) and (f). However, exclusively IR3 treatment generates completely residue-free fibers in CFRP A. Furthermore, the dominant damage mechanism through IR1 and IR3 treatment is fiber narrowing independent of the applied CFRP material. In contrast to that, UV1 causes straight fiber breakage on CFPR A and B as showed in Fig. 1 (f) and (h) (CFRP B surfaces look similar after IR1-3 and UV1 treatment, that is why they are left out in Fig. 1). In Fig. 1 (c) and (f) the SEM images show that laser radiation with the low-energy parameter sets IR2 und UV2 lead to partial matrix removal on top of the blank fibers with just occasional straight fiber breakages. Furthermore, UV2 treatment of CFRP A leads to partially embedded fibers, as showed in the greatly magnified image in the upper right corner of Fig. 1 (e). UV2 treatment of CFRP B leads to total removal of the epoxy matrix as well as straight fiber breakages (Fig. 1 (h)). That means that in this case irradiation with the UV2 parameter set is already sufficient to completely remove the matrix and uncover the carbon fibers.

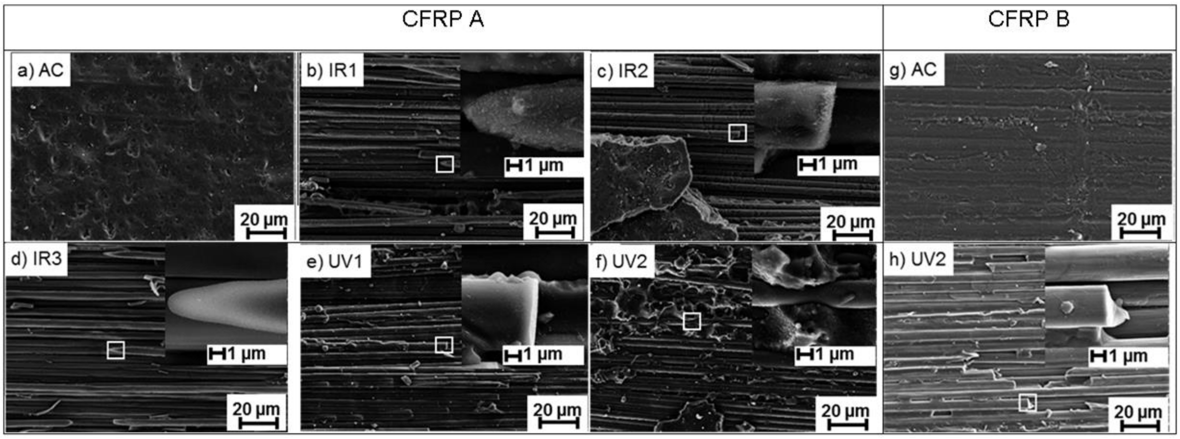

Fig. 1. Exemplary SEM images of representative areas after cleaning with acetone (a) and after treating with IR (b)-(d) or UV (e) and (f) of CFRP A as well as after acetone cleaning (g) and UV2 (h) for CFRP B. The fiber damage catalogue includes different kinds of defects like breakage, narrowing or oxidative degradation of the fiber. Higher magnified SEM images in the upper right corner illustrate the dominating fiber damage (b)-(f) and (h). (This illustration is published with permission of author Reitz et al. 2017)

To put it briefly, all parameter sets cause deboning for CFRP A except UV2 (see Fig. 2 (d)). With CFRP B (which is not showed here) debonding at the fiber-matrix interface is detected by any of the parameter sets even UV2, the set with the lowest intensity. These circumstances are displayed in Fig. 2.

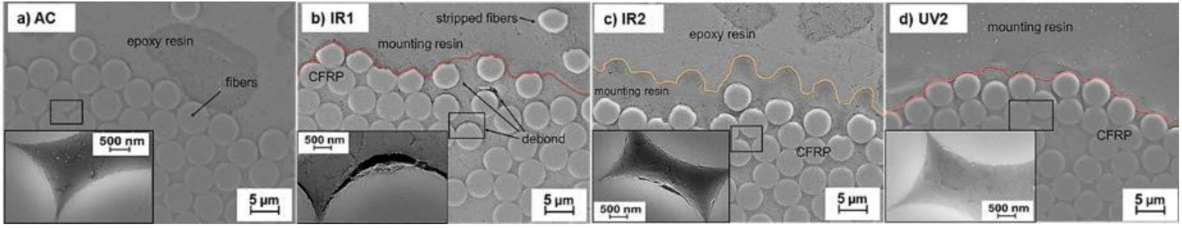

Fig. 2. SEM images of untreated (a) and laser pretreated (b)-(d) CFRP A. Debonding is generally observed after IR laser pretreatment (IR1, IR2 and IR3, which is not shown here) but not after UV2. (This illustration is published with permission of author Reitz et al. 2017)

So it is obvious that debonding is caused by laser treatment. Additionally, primary the fibers in the top layers are affected by the laser beam.

3.2. Shear strength of hybrid single-lap-joints

Table 2 displays the results of the mechanical tests, i.e. the average shear strength τ_s of single-lap joints for both CFRP A and B bonded to AlMg3. The shear strength of single-lap joints with CFRP A as adherend increases significantly after laser pretreatment with any types of described parameter sets. For CFRP B no increase of shear strength after treatment with any parameter could be detected. Further optimization has to be applied.

Table 2. Experimental shear strength τ_s of the SLJ tests.

Type of Material	**Unit**	**Parameter set**					
		AC	UV1	UV2	IR1	IR2	IR3
CFRP A	[MPa]	$8.2^{+0.5}_{-0.7}$	$12.5^{+0.5}_{-0.6}$	$14.9^{+0.9}_{-0.4}$	$12.1^{+1.2}_{-0.6}$	$12.2^{+0.5}_{-0.7}$	$15.6^{+4.1}_{-3.7}$
CFRP B		$19.1^{+1.7}_{-1.5}$	$12.3^{+0.4}_{-0.4}$	$16.6^{+1.2}_{-1.0}$	$13.7^{+1.1}_{-0.6}$	$16.8^{+0.7}_{-0.7}$	$13.5^{+0.4}_{-0.4}$

4. Discussion

4.1. Microstructural characterization

Different effects of the laser radiation on the surface structure and composition were detected in SEM images. Firstly, applying low-energy parameter sets (IR2 and UV2) led to an incomplete remove of the epoxy matrix, in comparison the high-energy parameter sets (IR1, IR3 and UV1) remove the matrix efficiently, however, the concentration of damaged fibers is increased as well. The effect of fiber narrowing, which was predominant observed after treatment with IR laser irradiation (see Fig. 1 (b) and (d)), can be explained by thermal degradation by oxidative decomposition that occurs in oxygen-bearing atmosphere above 600 °C (Minus et al. 2005). Both CFRP showed the same characteristic surface quality after IR laser treatment. It looks different by pretreatment with UV laser radiation. The reason for incomplete removal on CFRP A and complete removal of the matrix with fiber breakages on CFRP B with the low-energy parameter set UV2 might be the particular chemical composition rather than the manufacturing-related differences like thickness of the residual surface-polymer layer that leads to different interactions.

Secondly, SEM images of cross sections indicate that laser radiation damages the fiber-matrix interface through nanoscale debondings (see Fig. 2 (b) and (c)). Debonding for both CFRP A and B is generally observed after IR laser pretreatment. Additionally, it is observable after UV treatment for CFRP B, too. Due to, the low absorption coefficient of the epoxy matrix most of the energy is absorbed by the fibers (Takahashi et al. 2016). In contrast to the presented study Takahashi used an UV laser with 266 nm wavelength and additionally, the laser beam had contact to the cut surface, so cavities are achieved down to microscale.

4.2. Shear strength and fracture patterns:

The fracture analysis of CFRP surfaces shows adhesion failure on the CFRP side after cleaning with acetone. Nevertheless, it is probable that cleaning the surfaces with acetone increases the adhesion through removing impurities and humidity. Since no indication of cohesive substrate failure (CSF) can be found, it is reasonable to assume that acetone cleaning does not significantly affect near surface substrate strength.

Taking a look at the laser pretreatments, the combination of the parameter sets IR1 and IR2 to IR3 leads to the best surface conditions in these investigations. IR3 achieved the highest value for shear strength with 15.6 MPa. Based on the work of Schmutzler et al. it is possible to explain the positive impact on the shear strength with oxidation of carbon fibers (see Fig. 2 (d)) which leads to improved wetting behavior and formation of covalent bonds between fibers and adhesive (Schmutzler et al. 2014). However, IR laser application leads generally to CSF on the CFRP side, due to the decrease of the fiber-matrix adhesion in the near surface area. Simultaneously, a strength-increasing surface activation occurs. In this context, an extra explanation might be the infiltration of the cavities and therefore a partial restoration of laminate integrity.

With UV laser pretreatment the fracture behavior changes. By using UV2 parameter set just adhesion failure on the CFRP side can be observed at τ_S of 14.9 MPa. Obviously, surface activation is partially successful, but the degradation of the fiber-matrix adhesion does not occur yet. Still, the surface activation is not sufficient. The reason could be remaining epoxy, which shows poor adhesion forces with the adhesive. Additionally, the missing fiber narrowing is a hint for an insufficient oxidative activation of the fiber surfaces. At the end, we could not clarify whether the adhesion strength could be further increased without damaging the CFRP laminate. Using high-energy parameter set UV1 the error pattern changes to a mixture of AF and CSF on CFRP side, what is accompanied by a decrease of shear strength of about 15 %. That indicates that the UV-laser effects the CFRP A in the same way like IR radiation. This makes the further optimization an essential necessity.

For CFRP B the situation changes completely. Laser-pretreated samples do not reach the shear strength of the reference samples. Apparently, it is possible to clean and to activate the surface efficiently so that strong interactions of the matrix with the adhesive occur and lead to a strong adhesion. By means of fracture analysis both AF and CSF were detected on the CFRP side after applying laser treatment prior to bonding. That is applicable for all used laser parameter sets applied in this study, resulting in CSF for all laser pretreatment samples. The highest shear strength on CFRP B – approximately 16.5 MPa – was reached with the low-energy parameter sets IR2 and UV2. However, for CFRP B pretreatment with both IR and UV laser is not recommended.

5. Conclusion

For this study the impact on adhesive joining of two differently generated CFRP materials with aluminum was investigated. In order to create different surface conditions the CFRPs were pretreated with IR or UV laser radiation. As reference, the laminates were solely cleaned and activated with acetone. The resultant surface conditions were qualitatively characterized by means of optical methods and those were correlated with mechanical properties of adhesively joint hybrid SLJ (Al/ CFRP).

The following conclusion can be drawn:

- Wavelength and energy input can be seen as critical parameter for laser pretreatment because of their influence on the surface as well as the fiber-matrix adhesion – which in turn have an impact on bond strength and fracture behavior. Conditional on this parameter and the interaction with the used CFRP laser treatment can lead to two opposing effects: increasing of bonding strength through surface activation and decreasing of bonding strength through damages like fiber-matrix debonding.
- The success of laminate pretreatment strongly depends on the used laminate, i.e. fiber, fiber sizing, chemical composition of the matrix and its manufacturing process: Best results in regards to shear strength of the joints are obtained with specific IR and UV laser parameter sets for CFRP A and solely acetone cleaning for CFRP B.
- For CFRP A: IR laser radiation leads to surface activation but subsequently the fiber-matrix integrity got damaged. By using an UV laser, it is possible to avoid this damage. However, the enhancement of adhesion between CFRP and adhesive is limited and we reached comparable strength values to the IR laser system. It was worked out that bonding strength of adhesively bonded single-lap joints is a system property.
- Analyzing CFRP B after pretreatment with both IR and UV laser fiber matrix debonding was detected, which results in cohesion substrate failure for most of the tested samples. In contrast to CFRP A, solely acetone cleaning leads to the best results i.e. suitable combination of surface activation without affecting the integrity of the laminate.

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