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Improving the bond strength of metal-FRP-hybrids with thermal sprayed copper using pulsed laser-based processing approaches

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Abstract

Ablation processes during laser treatment of carbon fiber-reinforced plastics (CFRP) with pulsed lasers of various wavelengths and pulse durations are investigated. Three different surface pre-treatment strategies are used, including laser-roughening, selective matrix removal and laser micro structuring. Various ablation mechanisms, including evaporation and photochemical ablation are reported, depending on the employed laser source. Selected laser structured substrates were coated with copper by a wire arc spraying process. Bonding strengths up to 18.7 ± 2.0 MPa were achieved in shear tensile tests, by the combination of the roughening process and the micro-structuring approach. Consequently, the bonding strength could be increased over ~ 180 % compared to the common pre-treatment by mechanical blasting.

Keywords: Surface Functionalization; Pulsed Laser; Fiber-Reinforced Plastic; Metal-Plastic Hybrid; Coating Deposition

1. Introduction

Due to their high specific stiffness and strength, fiber-reinforced plastics (FRP) are ideal for the substitution of some metallic materials and thus reducing the weight of components (Schürmann, 2007). The use of FRP is limited by their physical properties and tribological behavior, including their low wear resistance (Qian et al.,

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2010) and insufficient thermal (Martins et al., 2018) and electrical conductivity (Zhao et al., 2019). Consequently, these typically metallic properties are still required on the FRP surfaces. Some of these constraints can be solved by surface metallization. Thermal spraying offers the possibility of applying thick coatings on a variety of materials to modify the surface properties (Lugscheider and Bach 2002).

In thermal spray processes, residual stresses mainly arise from the shrinkage of the spray particles after solidification and from differences of the thermal expansion coefficients from the coating and the substrate, which is given for the material combination of metal and FRP. (Takeuchi et al., 1990; Kuroda et al., 1988; Godoy et al., 2002; Schürmann, 2007). For this purpose, before the coating process, the surface of the FRP undergoes a pre-treatment process with the aim of roughening the surface. Thus facilitating mechanical interlocking between the coating material and the substrate. According to the state of the art, mechanical blasting is therefore used attributable to metal substrates (Gonzalez et al., 2016; Liu et al., 2006b; Liu et al., 2006a; Huonnic et al., 2010). Unfortunately, mechanical blasting on FRP leads to damages of the surface-near fibers due to the sharp-edged of the highly accelerated blasting particles (Liu et al., 2006b; Ramaswamy et al., 2020; Wingfield, 1993). This issue can be avoided by using laser pre-treatments as reported by different authors (Liebsch et al., 2019; Gustke et al., 2021; Gonzalez et al., 2016; Gebauer et al., 2019; Gebauer et al., 2018).

In addition, a deeper understanding of the ablation phenomena during the laser treatment can enable the development of application-specific surface structures, as has been pointed out in different studies (Akman et al., 2020, Kreling, 2015, Hart-Smith et al., 1996, Gebauer et al., 2020,).

This work, shows different surface pre-treatments based on latest investigations of Gebauer et al. [Gebauer et al., 2018, Gebauer et al., 2019, Gebauer et al., 2020], produced by different laser technologies. An enhancement of the adhesion strength of wire arc sprayed copper coating to CFRP substrate is investigated using the shear tensile test. Furthermore, a possible dependence of the bond strength on the roughness is examined.

2. Materials and Methods

2.1. Materials

A thermoset CFRP (SIGRAPREG® C U 600-0/SD-E501/33%) with 2 mm thickness consisting of an epoxy resin with unidirectional oriented carbon fibers (from SGL Carbon) with a fiber volume content of 67 % was used as substrate material. The used coating material was a copper wire (>99.8 % Cu) with 1.6 mm diameter (from GTV Verschleißschutz GmbH). To perform the shear tensile test, a metallic counterpart made out of an aluminum sheet (EN AW6082) was utilized.

2.2. Methods

Two laser systems with various wavelengths and pulse durations were chosen to generate different surface structures. The used laser sources consisted in a Nd:YAG laser with the wavelength of 355 nm and a pulse duration of 30 ns (denoted as short-pulse ultraviolet, SP-UV) to roughen the CFRP surface and to generate trench-like structures. Furthermore, a short-pulsed CO_2 laser with a wavelength of 10.6 µm (SP-IR-C) was used for selectively removal of the epoxy matrix. The technical specifications of the used lasers and their optical setups are presented in Table 1.

The mechanical blasting (MB) structure is used as a reference to the state of the art, produced under 2 bar pressure and an angle of 45 degree with corundum. The laser-roughening (R) by laser imitates the mechanical blasting structure while avoiding fiber damage. The selective matrix removal (SMR) should offer a high amount of enlarged surface area and undercuts, increasing the bonding strength (Habenicht, 2009). The combination of roughening with a trench-like structure (TS) also leads to a massively increased surface area compared to the state of the art. Detailed information about the geometrical design of the laser structures are listed in Table 2.

Table 1. Used laser systems including optical setups.

	SP-UV	SP-IR-C
Laser type	Nd:YAG	CO_2
Max. power in W	20	250
Wavelength in nm	355	10600
Pulse duration in s	$30 \cdot 10^{-9}$	$3 - 400 \cdot 10^{-6}$
Focal length in mm	160	200
Fluence in J/cm²	6.4-9.4	2.4

Table 2. Topographical parameters of produced structures.

Surface structure	Laser system	Hatch distance (µm)	Average trench depth (µm)
R	SP-UV	20	-
SMR	SP-IR-C	88	-
R+TS1	SP-UV	300	50
R+TS2	SP-UV	300	70
R+TS3	SP-UV	300	100

The copper coating was applied on the pre-treated CDRP using a wire arc spraying process, schematically shown in Figure 1. Compared to other thermal spraying processes, wire arc sprayed coatings exhibit improved mechanical properties (Lugscheider und Bach, 2002) and lead to a lower thermal impact into the FRP substrate (Cotell, 1999; Davis, 2004).

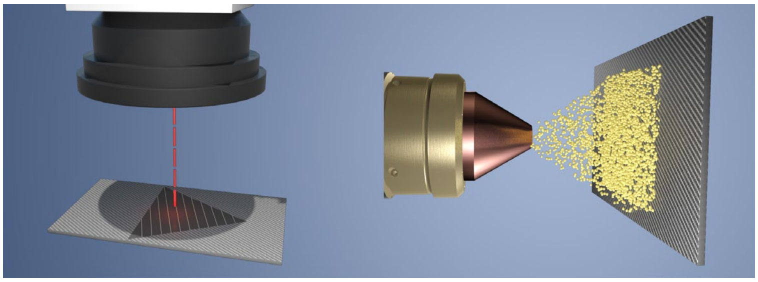

Fig. 1. Schematically process chain of laser structuring and the following thermal spraying process, © Fraunhofer IWS

The coated CFRP samples and the mechanical blasted metallic counterparts were joined using the FM 1000 adhesive film (from HTK Hamburg GmbH) to produce test samples for the shear tensile tests.

2.3. Characterization methods

The substrate surfaces were analyzed before the coating process using the laser scanning microscope (LSM) VK-X250K from Keyence. This contactless measurement is not affecting the filigree and contact-sensitive surface structures. Using the LSM, the substrate roughness S_a of all surface pre-treatments were measured over an area of 14 mm². Furthermore, using the scanning electron microscopy (SEM) JSM-6610LV from Jeol, the topography of the generated surface structures was analyzed in detail. The SEM operated in a working distance of 20 mm at 15 kV acceleration voltage.

The shear tensile strength of the copper coating was determined based on the DIN EN 1465, using the tensile and compression testing machine Zwick Z020 from Zwick/Roell, equipped with a 20 kN load cell. Six specimens were tested to verify the influence of each surface structure.

3. Results and Discussion

3.1. Surface structures

In Figure 2, SEM images of the pre-treated CFRP surfaces are shown. The mechanical blasted and the laser-roughened surfaces (in Figure 2a and 2b, respectively) show a highly irregular surface. Laser-roughening removes matrix material superficially, but without deep fiber exposure. In contrast, with mechanical blasting no fiber exposure was achieved and the surface appears considerably more fissured and uneven with a high amount of smashed fiber filaments. In addition, single corundum particles have been embedded in the soft plastic during the mechanical blasting process. The selective matrix removal with the SP-IR-C laser (Figure 2c) achieved a very uniform and deep exposure of the reinforcing fibers. Figure 2d exemplary shows the combination of laser-roughening and grid structure. Earlier investigations have shown, a full surface processing is necessary to avoid smooth, untreated surfaces between the trenches. Otherwise, untreated areas can lower the reproducibility of the adhesion strength or even avoid the wetting of the FRP by metal during the coating process. (Gebauer et al., 2018; Gebauer et al., 2019)

The treatment with the SP-UV laser only exposed the upper surface of the carbon fiber filaments (Figure 2b). Partly, the matrix material still covered the fibers. In the remaining epoxy matrix, single ablation traces of the laser beam can be observed. This traces validate a photochemical ablation mechanism, as was previously investigated by Akman et al. (Akman et al., 2020) and Kreling et al. (Kreling, 2015) for the treatment of plastic-based materials with ultraviolet laser radiation. Furthermore, the SEM images indicate a slight ablation of the fibers resulting in narrow grooves across the fibers (see Figure 2b). In addition to the photochemical ablation of the epoxy, there is also a strong interaction between the ultraviolet laser beam and the carbon fiber supporting the material ablation.

The matrix material was completely ablated giving rise to exposed fibers using the SP-IR-C laser (Figure 2c). Neither structural damage nor residues could be observed. Thus, the wavelength of 10.6 µm is ideal for the ablation of transparent materials such as the epoxy matrix. Additionally, the thermal conductivity of carbon fibers, which is 4.9 W/mK along the fibers and only 1.7 W/mK in the perpendicular direction (Schürmann, 2007) leads to heating of the material following mainly the fiber direction. In combination with the comparatively long pulses of the SP-IR-C laser, the laser process led to a strong heating of the matrix material and the reinforcing fibers. Consequently, the epoxy material could be completely removed by evaporation, a photothermal ablation mechanism as explained by Akman et al. (Akman et al., 2020)

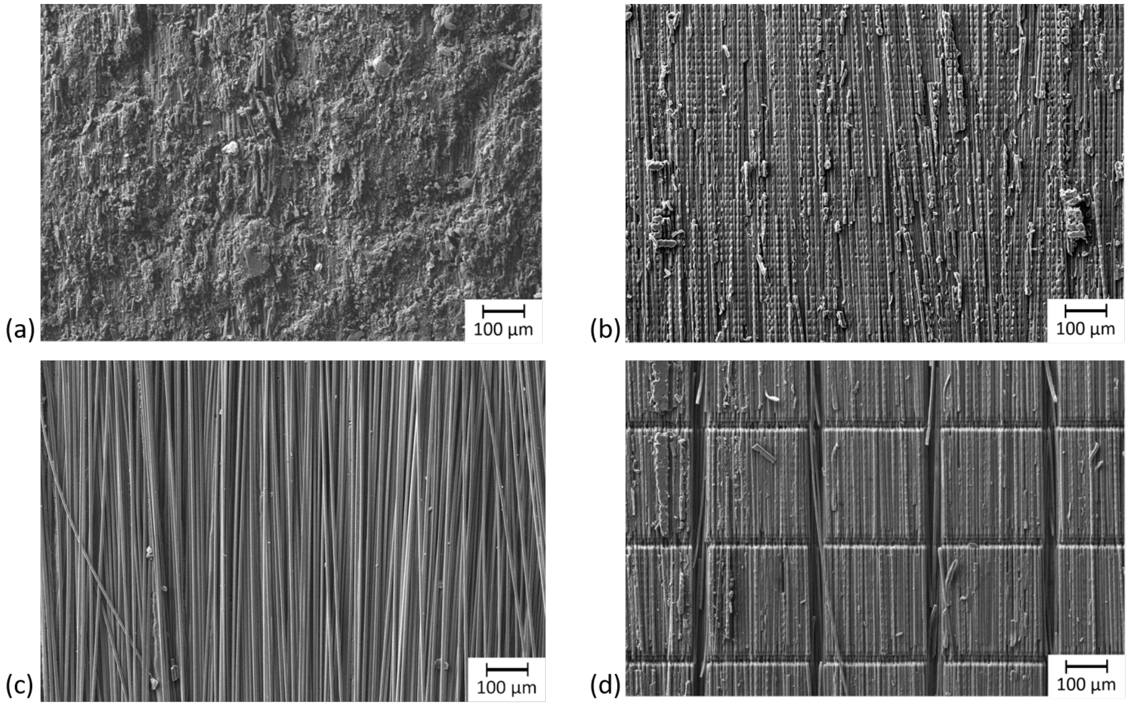

Fig. 2. SEM images of (a) Mechanical blasting; (b) Laser-roughening; (c) Selective matrix removal; (d) Laser-roughening plus trench-like structure with 399 µm trench distance and 70 µm trench depth [Gustke et al., 2021].

3.2. Adhesion strength

Figure 3 shows the adhesion strength of the copper-coated CFRP from the shear tensile test for each surface condition, supplemented by the roughness values S_a of the CFRP before thermal spraying. Although the laser-roughening structure has a significantly lower roughness of 5.5 µm than the mechanical blasting sample with 13.5 µm, the two structures achieve similar adhesion strengths of 11.1 ± 2.5 MPa and 10.0 ± 0.9 MPa, respectively. A similar adhesion strength of 10.5 ± 0.9 MPa could be achieved with the selective matrix removal structure with a roughness S_a of 9.3 µm. Only the combination of laser-roughening and trench-like structure lead to a significant increase of the adhesion strength. With increasing trench depth the adhesion strength of the copper coating to the laser-treated CFRP substrate was rising from 13.6 ± 1.4 MPa over 14.3 ± 1.0 MPa up to 18.2 ± 2.1 MPa for 50 µm, 70 µm and 100 µm trench depth respectively. Furthermore, the roughness of the samples was rising with increasing trench depth from 5.9 µm and 12.7 µm up to 15.1 µm, respectively.

The samples with the lowest trench depth of 50 µm present the lowest substrate roughness and the lowest adhesion strength, comparing the three combined laser structures with each other. The order of the substrate roughness values of the different trench-like structured samples relates to the order of their achieved coating adhesion strength. However, this correlation was not observed when comparing the different structure types. Consequently, it can be determined that the adhesion strength of the thermal sprayed coating to the substrate does not directly depend on the roughness value S_a but also on the texture geometry. Clearly, the clamping effect of the shrinking coating into the trenches enable much higher adhesion strengths compared to the roughening by mechanical blasting despite a similar roughness around 13 µm.

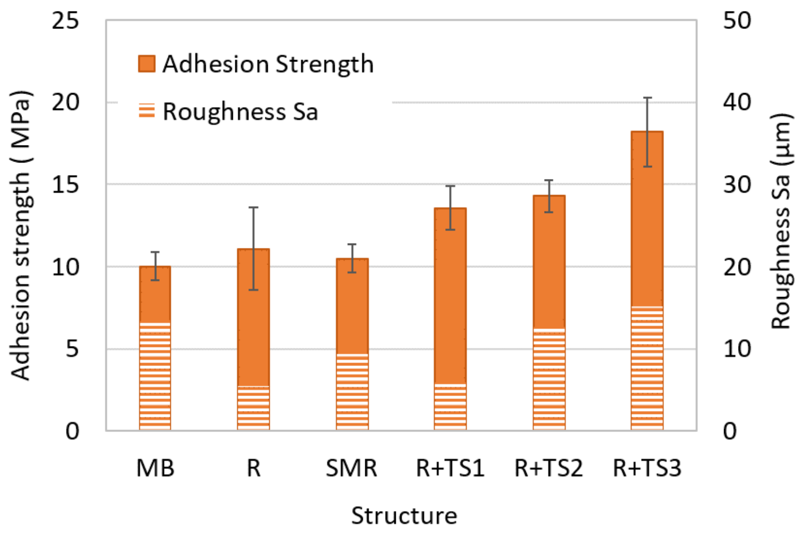

Fig. 3. Adhesion strength and roughness S_a for all surface structures

4. Conclusions

It was shown, that laser radiation on CFRP leads to different ablation mechanism depending on wavelength and pulse duration. Due to the high thermal conductivity of carbon fibers in longitudinal direction, material removal is significantly supported, for example by increased evaporation of the plastic outside the interaction zone of the SP-IR-C laser beam with the FRP. To counteract the shrinkage of the metal spray particles after solidification on the one side and to compensate the differences of the thermal expansion coefficients from the metal coating and the FRP-substrate on the other side, a rough surface is not sufficient to enable a reliable bond strength. The results have shown a high potential of all laser structured samples compared to the mechanical blasted reference. The average shear tensile strength of the laser-structured CFRP range between 111 % and 182 % compared to the mechanical blasted samples. The combination of laser-roughening and a trench-like structure proved to be particularly suitable. With a trench distance of 300 µm and a trench depth of 100 µm adhesion strength up to 18.2 MPa could be achieved.

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