FUSION BONDING OF FIBER REINFORCED THERMOPLASTICS AND THERMOSETS

H.Grefe[1], S.Kreling[1], R.van Moorleghem[2], I.F.Villegas[2] and K.Dilger[1]

[1]Institute of Joining and Welding, Composite Technologies, Technical University of Braunschweig, Langer Kamp 8, 38106 Braunschweig, Germany
Email: h.grefe@tu-braunschweig.de, Web Page: http://www.ifs.tu-braunschweig.de
[2]Aerospace Structures and Materials Department, Delft University of Technology, Kluyverweg 1, 2629 HS Delft, The Netherlands
Email: I.FernandezVillegas@tudelft.nl, Web Page: http://www.lr.tudelft.nl

Keywords: composite welding, fusion bonding, thermoset composite, thermoplastic composite

Abstract

Fusion bonding of thermoplastic parts will most likely result in joints strengths, close to the origin cohesive strength of the matrix material, if suitable parameters are applied. However, fusion bonding of thermoplastic materials and thermosets will generate an adhesion based joint, with the strength highly dependent on the thermosets surfaces conditions.
In this paper different surface pretreatments were investigated regarding the application on hand. The carbon-fibers of an aerospace epoxy prepreg were exposed using different laser parameters also the surface of an automotive RTM material was grit blasted and also treated with a laser. The thermosets afterwards were joined to their appropriate thermoplastic composite using adhesive bonding, conductive or ultrasonic welding.
The laser pretreated surfaces were analyzed using SEM, lap shear samples were welded and tested and the joints were examined using cross section micrographs.

1. Introduction

The vast use and nearly endless variability of fiber reinforced materials, which are used nowadays, is clearly the best argument in regard of their advantages as a construction material. A suitable combination of fiber and matrix materials can be found for nearly any kind of application.
On the grounds of this variety of different materials, their properties and behavior, the pursuit for a *well suited* joining technique is getting more and more complex.
In most cases mechanical joining and fiber reinforced composites are not seen as an ideal arrangement. Delamination due to the drilling, concentrated stresses caused by the holes and furthermore downsides disqualify mechanical joining for a lot of composite applications [1]. On the other hand, in contrast to adhesive bonding no surface preparation is required, the process is well known and the necessary tools are rather ordinary. In general, choosing an appropriate joining method is mostly a compromise based on the weighting of requirements.
The possibly most crucial factor for non-mechanical joining of fiber reinforced plastics (FRP) is the matrix material. The major market volume is taken by thermoset epoxy-resins, processed through different production methods e.g. autoclave curing in aerospace applications or resin transfer molding (RTM) for automotive structures. Nonetheless thermoplastic matrix materials are gaining market shares as well [2]. Thermoplastic composites offer a good damage tolerance with a high impact resistance and fracture toughness. Also the consistency against solvents and corrosion, the fatigue behavior, the virtual infinite shelf life and the ability to be reformed, repaired and reprocessed are their main benefits. Furthermore, apart from the classic joining techniques thermoplastic materials are also

Excerpt from ISBN 978-3-00-053387-7

suitable for fusion bonding. This introduces a variety of bonding methods usually divided by their heating mechanism or energy source [3]. Welding and fusion bonding of thermoplastic materials is widely used in commercial applications and the process is consequently well understood. Another joining method, operating on -at least- related physical principals is hot melt adhesive bonding. This broadly established technique uses thermoplastic adhesives which are applied onto the adherends in a liquefied state and form a joint during solidification which might also be assisted by crosslinking.
Including a thermoplastic adhesive as a functional layer in the thermoset adherend [4] would cut the necessary application of adhesives out of the joining process. A severe drawback to this technique is the additional effort of producing or including a functional layer in the thermoset part. Fiber reinforced thermoplastics however do not necessarily require additional layers for fusion bonding. The thermoplastic matrix could function as adhesive material. This is also possible in regard of joining fiber reinforced thermoplastics with other materials, e.g. metallic components [5].
The paper on hand investigates the technical feasibility of directly joining fiber reinforced thermoplastics and thermosets without the use of additional adhesive. Two different material combinations were used: an automotive relevant combination of glass-fiber reinforced polyamide 6 (PA6) joined with an RTM manufactured carbon-fiber reinforced epoxy and a carbon-fiber reinforced polyether ether ketone (PEEK) combined with an autoclave cured carbon-fiber reinforced epoxy prepreg.
Fusion bonding as a joining technique for fiber reinforced thermoplastics has a simple benefit: due to the complete melting of the surface and interface layers, the challenge in adhesive bonding due to the comparably low surface energy of the thermoplastic matrices is thereby avoided. The strength of the joint can even achieve the values of the bulk material [6, 7].
The joining processes should fit into a possible serial production with the desire to be faster than regular adhesive bonding while hopefully achieving a higher joint strength or reducing the process effort. During the process, pretreatment of the thermoplastic surface can be omitted, adhesive neither has to be applied nor cured. The still necessary pretreatment of the thermoset adherends is investigated in the following.

2. Experimental

In order to directly fusion bond or weld thermoplastic to thermoset composites, without the use of a thermoplastic functional layer on the thermoset composite, two different types of surface treatments were explored: sandblasting and laser treatment, which are fast-and-easy to automate processes. These surface treatments were applied to the thermoset composite with the idea of promoting mechanical interlocking between the molten thermoplastic resin and the thermoset composite during the welding process. Two different welding processes, conduction and ultrasonic welding, were evaluated together with adhesive bonding as a reference joining process. The details on the experimental work performed in this research are provided below.

2.1. Materials

To examine the feasibility of fusion bonding, two material combinations were used regarding different fields of application. A typical automotive material combination was investigated using woven glass fiber reinforced polyamide 6 and quasi-isotropic non-crimp carbon fiber reinforced epoxy, which was fabricated in a resin transfer molding process. This material combination was joined using conduction welding and adhesive bonding as a reference.
As typical aerospace material combination, a woven (5 harness satin) carbon fiber reinforced PEEK (CETEX, Ten Cate Advanced Composites) material was chosen to be joined with epoxy reinforced with woven (twill weave) carbon fibers (Hexply M18/1, Hexcel Composites). $[0/90]_{3S}$ CF/PEEK laminates with a final thickness about 2 mm were manufactured in a hot plate press at 385 °C and 10 bar for 20 min. $[0/90]_{4S}$ CF/M18 laminates were manufactured in an autoclave at a curing temperature

Excerpt from ISBN 978-3-00-053387-7

of 180°C and 2 hours curing time, following the instructions of the manufacturer. In this case the joining was performed using two different techniques, conductive welding and ultrasonic welding.

2.2. Pre-Treatment

For the laser pre-treatment an ns-pulsed third harmonic solid state laser, emitting radiation of 355 nm wavelength was used due to good results in previous studies. As a result of the decent absorption, the high photon energy and the comparably low thermal influence of the UV radiation a removal of the matrix resin without thermal degradation of the bulk material can be achieved. [8]
For the grit blasting aluminum oxide was used. The blasting pressure was kept rather low, so it would only penetrate the resin surface and not to damage the fibers underneath.

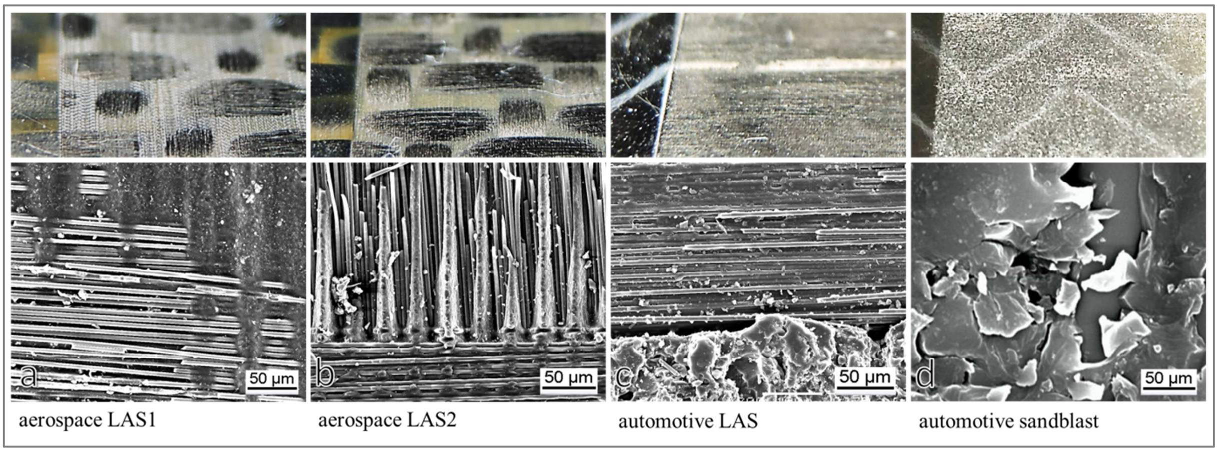

Figure 1. Photos and electron microscope images of the treated surface.

As shown in figure 1 the aerospace material was treated with two different parameters. Configuration LAS1 (a) only partially removed the surface resin and caused only minor damage to the fiber. Whereas configuration LAS2 (b) removed the largest part of the surface resin and exposed most of the accessible fibers. Due to the more aggressive treatment more fibers have been damaged and the rather deep penetration of the laser beam might lead to delamination.
The automotive composite, due to its production process, has a much thicker surface resin layer. Therefore more resin had to be removed to expose the fibers and more energy was introduced into the material. This caused a more severe abrasion and also more loose fibers which had been cleared from the resin. Also the non-crimp fiber design generally includes a stitching yarn, which separates the rovings and creates a fiber free trail along the fiber orientation.
The low pressure grit blasting broke open the surface layer as intended: enlarging the effective surface area, removing auxiliary processing materials and enabling mechanical interlocking. No damage to the fibers was detected, only the resin layer has been penetrated.

2.3. Joining methods

Ultrasonic welding
Individual test samples were ultrasonically welded in a single lap configuration with 12.7 mm overlap using a Rynco Dynamic 3000 ultrasonic welder and a welding jig custom-designed and built at TU Delft (Fig. x). A displacement-controlled welding process was used with the following welding parameters: 1500 N welding force, 86.2 µm peak-to-peak amplitude, 0.10 mm travel (i.e. displacement

Excerpt from ISBN 978-3-00-053387-7

during vibration), 1000 N solidification force and 4 s solidification time. The travel value was chosen based on the feedback provided by the ultrasonic welder following the procedure explained in [9]. 0.25 mm-thick flat energy directors, i.e. a layer of neat PEEK resin, were used during the welding process to effectively concentrate heat generation at the welding interface [10]. The welding parameters were chosen to ensure heating times below 500 ms to avoid thermal degradation of the epoxy resin as explained in [11].

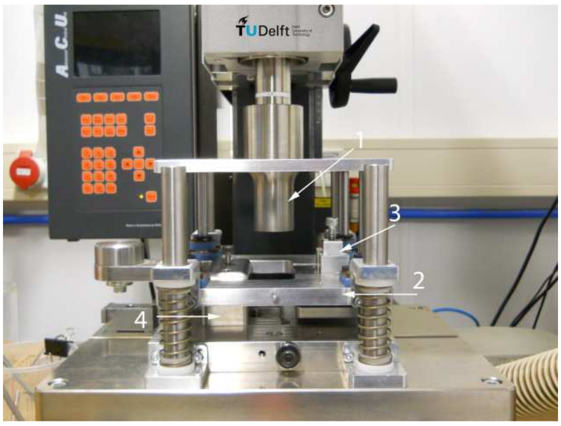

Figure 2. Ultrasonic welding equipment and clamping jig. (1) Sonotrode, (2) sliding platform for clamping of top adherend, (3) clamp top adherend, (4) clamp bottom adherend.

Conductive welding

The joint is placed between two conductive heating elements, which are embedded in a metal casing. Both parts are held in place by the joining pressure. The joining process is not regulated by the displacement but through the force applied by the cylinders. Therefore the actual displacement may vary slightly between the samples. The joining force for both material combinations was set to 16 N for an overlap of 15 x 25 mm. The temperature of the lower and the upper heating strip is regulated independently.

Due to the individual material properties, different joining strategies have been employed. Completely melting the thermoplastic material consumes a lot of time and energy. Therefore the automotive GF/PA6 was only heated up to 195 °C and kept well under its melting point. The corresponding CF/RTM material is able to withstand a temperature of 320 °C for a short time. While under joining pressure, it is heated up to 260 °C to melt the thermoplastic material in the joining area and thereby form the bond. Keeping the PA6 under its melting point (220 °C) reduces the amount of molten material and the risk of pores and cavities in the joint. Also a better surface quality on the non-welded side is sustained. The joint displacement or respectively the compression of the thermoplastic material during the joining process was measured with an average of 0.3 mm.

To melt the PEEK matrix of the aerospace material combination a higher temperature is required (335 °C). Even though the aerospace epoxy has a higher temperature tolerance compared to the automotive matrix, the required temperature is too high to press the overheated epoxy into the thermoplastic matrix. Therefore the PEEK is molten (heating temperature 370 °C), and pressed onto the warmed epoxy (290 °C). The average compression was measured at 0.2 mm.

Excerpt from ISBN 978-3-00-053387-7

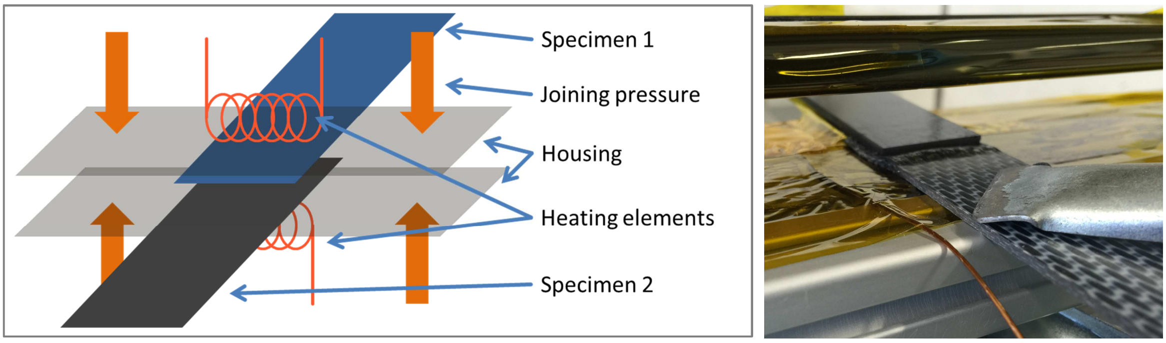

Figure 3. Conductive joining device and PEEK specimen.

2.4 Testing and analysis

The ultrasonically welded samples were tested in a Zwick Roell 250KN universal test bench following ASTM D 1002 standard. The cross-head speed was set at 0.13mm/min and the clamps were offset to ensure load application parallel to the weldline. Five samples were welded per set of parameters.
To test the conductive welded specimen, a Instron 5566 with a 10 kN load cell was used. The testing was performed following the DIN EN 1465 (which is quite comparable to the ASTM D 1002).
Cross section microscopy and fractography were used as well to gain deeper understanding on the output of the different welding processes considered in this study.

3. Results and discussion

Lap shear strength
Comparing the lap shear results of the different joints (figure 4); some general observations can be made: Regarding the aerospace combination, ultrasonic welding achieved a higher lap shear strength; whereas the highest overall strength was attained by the conductively welded automotive joints.

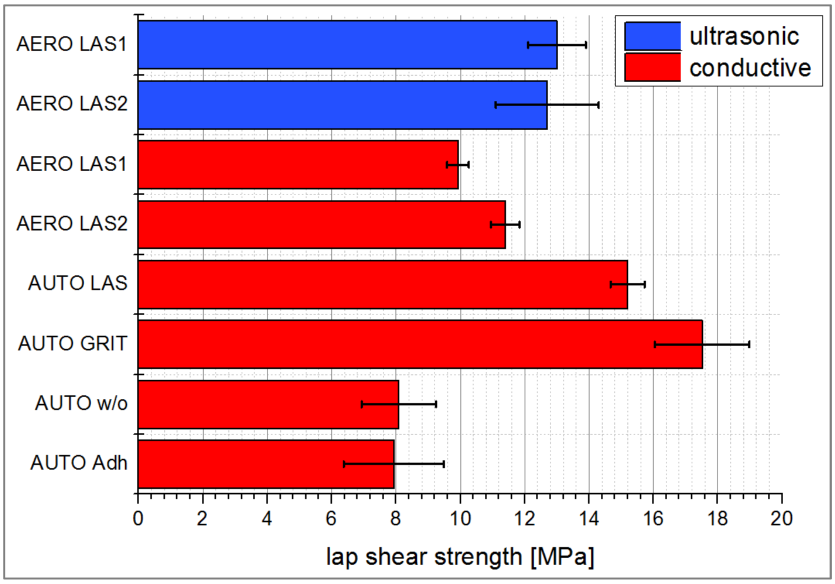

Figure 4. Results of the lap shear testing.

Excerpt from ISBN 978-3-00-053387-7

Comparing conductive or adhesive bonding without any surface pretreatment (figure 4 AUTO w/o and AUTO Adh) the grit blasted conductively welded PA6 – RTM joints had double the LSS.
Regarding the different material combinations joined by conductive heating the laser treated automotive combination (figure 4 AUTO LAS) has a 40% higher LSS than either of the conduction joined aerospace materials (figure 4 AERO LAS1 and AERO LAS2). The lap shear strength of the grit blasted joints is even 60% higher.
Considering the material properties of the thermoplastic matrices or respectively the adhesives, a significantly higher LSS would have been expected of the aerospace combination.

Failure patterns
The lower than expected LSS of the aerospace material combination might be caused by the different fiber volume content of the composites. Even though the PA6 was only partially molten, it had a larger joining compression than the PEEK composite which had been completely liquefied. The PEEK joint was thereby comparatively dry with only little matrix to bond to the exposed carbon-fibers. Considering the failure modes (figure 5) supports this assumption.

Figure 5. Fracture patterns of welded specimen (AUTO GRIT, AUTO LAS, AERO LAS1).

The laser treated RTM-specimens all failed by delamination of the upper fibers which were joined to the thermoplastic matrix.
The grit blasted samples show a separation of the surface resin film from the topmost fiber layer. A higher LSS would have been expected by the laser treated specimen, due to the direct bonding onto the fibers, however the production process of the RTM material created a rather thick resin surface film. To remove this, an intense laser treatment was necessary. This causes in delamination due to thermal degradation of the residual matrix (figure 6) [12]. In other studies, using the same thermoplastic material, the same joining process and equipment for a non-crimp epoxy prepreg with no pretreatment at all a LSS of 14 MPa has been reached.

Excerpt from ISBN 978-3-00-053387-7

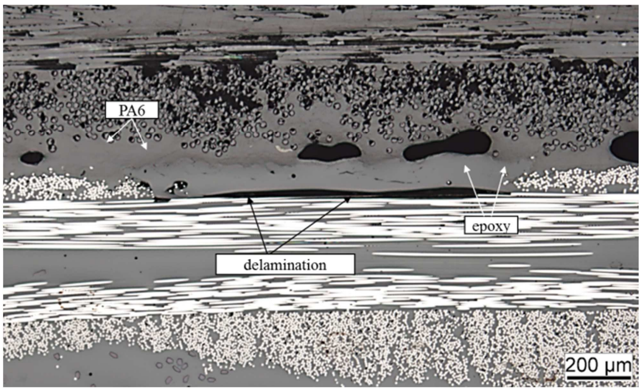

Figure 6. Cross section of the delamination in the RTM epoxy.

Cross sections

The cross section micrographs shown in figure 7 for ultrasonically welded samples with LAS1 and LAS2 laser treatment, respectively, show that despite the very short welding times, excellent wetting of the laser-treated epoxy surface by the PEEK resin was achieved. They also confirm the absence of thermal degradation in the epoxy adherends, which would cause resin sublimation [11] and result in porosity in the epoxy adherend. However, the LSS values obtained for the CF/PEEK-CF/epoxy welded joints were significantly lower than values obtained for similar material combinations welded through a thermoplastic resin layer co-cured to the CF/epoxy composite [13]. This results from the fact that in this research adhesion between the thermoplastic and laser-treated CF/epoxy relies solely on mechanical interlocking. More intricate textures on the surface of the CF/epoxy composite could hence be expected to result in higher weld strengths.

The conductive welded CF/PEEK – CF/M18 showed a similar behavior. The cross section of the LAS1 parameter (see Figure 7) shows isolated fibers, which are partially still bound to the epoxy resin but mostly embedded in the PEEK pressed onto them. The overview shows some defects in both adherends. Inside the CF/PEEK part, some minor pores are visible, which possibly already occur in the base material. The CF/M18 shows some interlaminar cracks which might be the first signs of thermal degradation due to the heat conduction. Further investigations are planned to clarify the cause of this defects. These defects might be the cause for the lower LSS which had been observed earlier (figure 4).

Excerpt from ISBN 978-3-00-053387-7

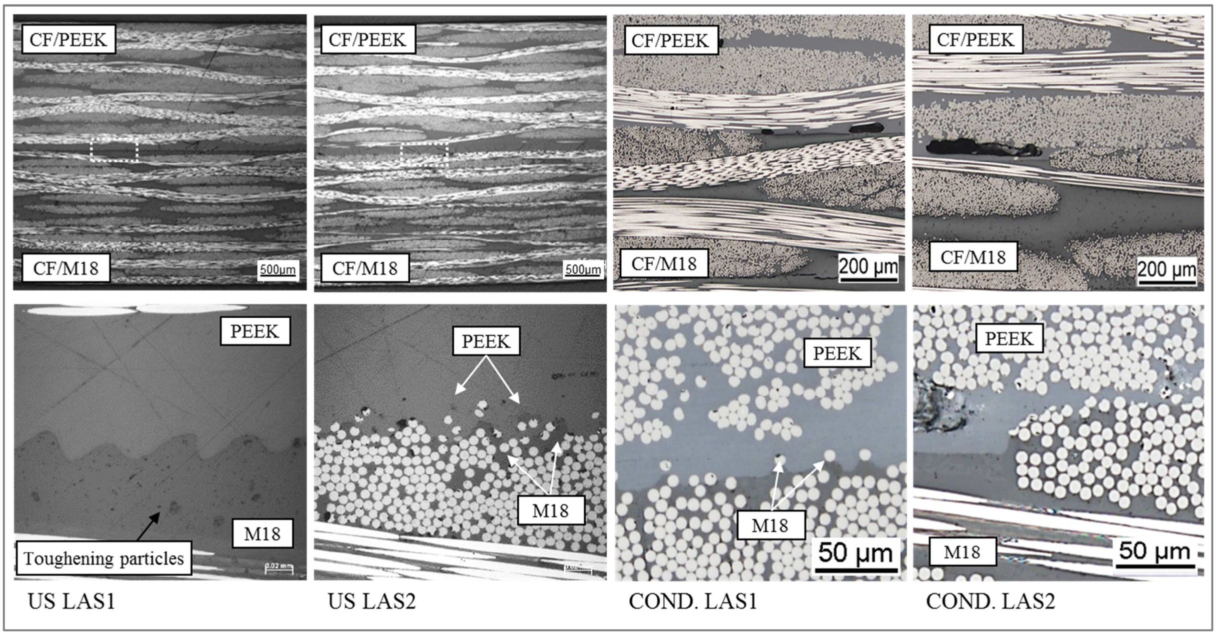

Figure 7. Cross section micrographs for a CF/PEEK – CF/M18, with PEEK resin (light grey) and epoxy resin partially removed by the laser treatment (darker grey).

The image of the LAS2 parameter (Figure 7) shows the result of the more intense treatment: the topmost fibers are locally damaged and partially removed. Due to the undulation a bigger ablation depth into the resin was necessary to expose a larger area of fibers. To accomplish this, the described local fiber deterioration had to be taken into account. The overview (figure 7 top, right) also shows a cavity, which is completely enclosed in thermoplastic material. So this void -like many other- is not part of the boundary layer but rather a pore inside of the thermoplastic composite.
For both conduction-welded configurations some interfacial voids occur, which presumably result from air, which is entrapped between the adherends and cannot effuse during the welding process.

The different laser parameters, used to pretreat the surfaces show more or less exposed fibers and accordingly residual resin. This causes different bonding conditions; the LAS2 parameter has an overall larger area of exposed fiber to bond on, whereas the treatment using the LAS1 parameter results in significantly more residual resin to bond on. The slightly higher LSS of the conductively bonded LAS2 specimens might be caused by a better adhesion of the PEEK to the fibers compared to the matrix material. To determine whether the amount of exposed fibers and residual resin is a major influence to the joint strength, further testing is planed with a non-crimp prepreg epoxy material which allows a complete fiber exposure through laser treatment.

4. Conclusion

Both material combinations applied in the present investigations show the general ability to be directly joined using the thermoplastic matrices as adhesive material. The necessary high temperatures of the conductive joining process for the aerospace material combination might result in thermal degradation and will therefore be further investigated. No thermal degradation, however, was found in the ultrasonic welded samples, which was probably the cause of higher lap shear strengths. Nevertheless the mechanical interlocking created between the thermoplastic resin and the thermoset composite was not enough to achieve the lap shear strength values yielded by ultrasonically welded samples in which a thermoplastic functional film was added to the thermoset composite [13].
As a continuation of this work also fusion bonding of thermoset/thermoplastic material combinations

Excerpt from ISBN 978-3-00-053387-7

using inductive heating of the carbon fibers will be investigated. Furthermore the origin of the pores and cavities which occurred in the conductively welded specimen will be explored.
Due to the delamination in the RTM-material caused by the laser pretreatment further testing will be conducted using alternative thermoset materials and also different pretreatment methods.

References

[1] H. Schürmann. *Konstruieren mit Faser-Kunststoff-Verbunden.* Springer-Verlag Berlin Heidelberg, 2007.
[2] E. Witten, T. Kraus, M. Kühnel. Composites-Marktbericht 2015. AVK – Industrievereinigung Verstärkte Kunststoffe e.V., Frankfurt, 2016
[3] A. Yousefpour, M. Hojjati and J.-P. Immarigeon. Fusion Bonding/Welding of Thermoplastic Composites. Journal of THERMOPLASTIC COMPOSITE MATERIALS, 17(4): 303-341, 2004.
[4] S. Denga, L. Djukicb, R. Patonb, L. Yea. Thermoplastic–epoxy interactions and their potential applications in joining composite structures – A review. Composites Part A: Applied Science and Manufacturing, 68: 121-132, 2015.
[5] K.Lippky, L. Gerling, F.Fischer and K.Dilger. Investigation of shear strength properties of fusion bonded lap-shear joints underthe influence of surface contaminations from deep drawing oils. Proceedings of the 37th Annual Meeting of the Adhesion Society, San Diego, CA United States, February 23-26, 2014
[6] M. Griese, D. Blass, E. Stammen and K. Dilger. Influence of laser pre-treatment on the mechanical properties of adhesively bonded CF-PEEK joints and their comparison to induction welded joints. Proceedings of the 39th Annual Meeting of the Adhesion Society, San Antonio, TX United States, February 21-24, 2016
[7] C. Ageorges, L. Ye, M. Hou, Advances in fusion bonding techniques for joining thermoplastic matrix composites: a review, Composites: Part A: Applied Science and Manufacturing, 32(6): 839-587, 2001
[8] F. Fischer, S. Kreling, M. Frauenhofer, and K. Dilger. Laser surface pre-treatment of CFRP for adhesive bonding in consideration of the absorption behavior. The Journal of Adhesion, 88(4-6): 350-363, 2012.
[9] I.F. Villegas. Strength development versus process data in ultrasonic welding of thermoplastic composites with flat energy directors and its application to the definition of optimum processing parameters. Composites Part A: Applied Science and Manufacturing, 65: 27-37 (2015).
[10] I.F. Villegas, et al. A comparative evaluation between flat and traditional energy directors for ultrasonic welding of CF/PPS thermoplastic composites. Composite Interfaces, 22(8): 717-729 (2015).
[11] I.F. Villegas and P. Vizcaino Rubio. On avoiding thermal degradation during welding of high-performance thermoplastic composite to thermoset composites. Composites Part A: Applied Science and Manufacturing, 77: 172-180 (2015).
[12] D. Blass, S. Kreling. S. Nyga. T. Westphalen, B. Jungbluth, et al. CFRP bonding pre-treatment with laser radiation of 3µm wavelength: laser/material interaction. Proceedings SPIE 9736, Laser-based Micro- and Nanoprocessing X, 973614, San Francisco, CA, United States, March 4, 2016.
[13] I.F. Villegas and P. Vizcaino Rubio. High-temperature hybrid welding of thermoplastic (CF/PEEK) to thermoset (CF/epoxy) composites. Proceedings of the 20th International Conference on Composite Materials, Copenhagen, Denmark, July 19-24, 2015.

Excerpt from ISBN 978-3-00-053387-7