HIGH-TEMPERATURE HYBRID WELDING OF THERMOPLASTIC (CF/PEEK) TO THERMOSET (CF/EPOXY) COMPOSITES

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ABSTRACT

Thermoset composites are widely used for the manufacturing of modern composite aircrafts. The use of thermoplastic composites (TPC) in aerospace applications is, however, gradually increasing owing to their cost-effectiveness in manufacturing and improved damage tolerance. An example of the use of thermoplastic composites in aircraft is the thousands of clips and cleats that connect structural elements in the fuselage of new composite passenger aircrafts, which are press-formed from pre-consolidated laminates in only a few minutes. Thermoplastic composite and thermoset composite parts are currently joined through mechanical fastening, which is known not to be an optimal joining technique for composites. Nevertheless, the ability of TPC to be welded with little surface preparation and short assembly times poses the question of whether thermoplastic and thermoset composites can be welded together. Hybrid thermoplastic to thermoset composite welding poses two main challenges: firstly, adhesion between the thermoplastic and thermoset resins; secondly, degradation of the thermoset resin when exposed to the welding temperatures. This paper presents a procedure to successfully prevent any negative thermal effects on the thermoset resin during high-temperature welding of thermoplastic to thermoset composites. The procedure is based on reducing the heating time to fractions of a second during the welding process. In order to achieve such short heating times, which are much too short for commercial welding techniques such as resistance or induction welding, ultrasonic welding is used. A particularly challenging scenario is analysed by considering welding of carbon-fibre reinforced poly-ether-ether-ketone (CF/PEEK), with a melting temperature of 340°C, to carbon-fibre reinforced epoxy (CF/epoxy) with a glass transition temperature of 157°C.

1 INTRODUCTION

Thermoplastic composites (TPC) are very attractive to several industries as a result of their cost-effective manufacturing and cost-effective assembling through welding. One of the most cost-effective manufacturing techniques applied to thermoplastic composites is press forming. Press forming allows to shape an initially flat thermoplastic composite laminate into a near to neat-shape part in several minutes [1]. In the aerospace industry, press forming is widely used for the manufacturing of small to medium-sized mass-high-volume-production thermoplastic composite parts. An example of this are the thermoplastic composite clips used for the fuselages of the new composite passenger aircrafts, Airbus A350 and Boeing 787 [2]. The manufacturing of bigger components such as fuselage or wing sections or panels relies, however, on the use of, more traditional, thermoset composites (TSC), which currently show cost advantages as compared to thermoplastic composites. The usage of both thermoplastic and thermoset composites for optimal manufacturing of different parts in the aircraft requires assembling of dissimilar TPC/TSC structures, which nowadays is solved through mechanical fastening [2]. Since TPC can be welded with little surface preparation and short assembly times, the possibility of applying thermoplastic welding technologies to the joining of dissimilar TPC/TSC structures, referred to in this paper as “hybrid TPC/TSC welding” is attracting the attention of the scientific community. Reliable hybrid TPC/TSC welding processes are expected to offer a significant reduction in assembly times, assembly costs and weight as compared to mechanical fastening.

One of two major challenges in welding of TPC/TSC structures is the adhesion between the thermoplastic and the thermoset composites. Researchers seem to agree on the necessity of coating the thermoset composite with a thermoplastic-rich layer through a co-curing process in order to achieve sufficient adhesion between the thermoset and the thermoplastic polymer prior to the welding process [3]. The thermoplastic coated thermoset composite parts are then either welded together or welded to thermoplastic composite parts by locally melting the thermoplastic/thermoplastic interface. Adhesion between the thermoset composite and the thermoplastic-rich coating layer can be based on micro- and/or macro-mechanical interlocking. On one hand, micro-mechanical interlocking entails the migration of thermoset pre-polymer molecules into the thermoplastic to create a semi-interpenetrating network. Successful creation of semi-interpenetrating networks has been reported for several combinations of compatible thermosetting and amorphous thermoplastic polymers, such as epoxy with polysulfone (PS) or polyethersulphone (PES), and bismaleimide (BMI) with PS, PES or polyetherimide (PEI) [4]. In these cases, the amorphous nature of the thermoplastic polymers and, therefore, their low resistance to solvents allows the uncured components of the thermoset component to migrate through the interface. Moreover, Hou et al state in [5] that combinations of semi-crystalline thermoplastics and thermosets with matching solubility levels, such as polyvinilidene fluoride (PVDF) and epoxy can as well generate semi-interpenetrating networks. On the other hand, macro-mechanical interlocking can be achieved through the co-curing of a composite hybrid layer partially impregnated with thermoplastic resin onto the thermoset composite laminate, as proposed by Jacaruso et al in [6].

The second major challenge in hybrid TPC/TSC welding is thermal degradation of the thermoset composite when exposed to the relatively high temperatures generated during the welding process. Degradation of the thermoset composite adherend during the welding process can be avoided through the selection of a thermoplastic/thermoset combination such that the welding temperature is lower or similar to the glass transition temperature of the thermoset resin. Such is the case of PVDF to epoxy welding. PVDF has a melting temperature of 170°C and therefore, it allows to perform the welding process at a temperature that does not cause severe overheating of epoxy composites cured either at 177°C or 127°C [5,7]. This method, however, greatly limits the nature of thermoplastic composites that can be welded to thermoset composites. When the application requires the use of high-performance thermoplastic composites, such as PEEK, polyether ketone ketone (PEKK), PEI or polyphenylene sulphide (PPS), with processing temperatures above 300°C, alternative solutions for ”high-temperature hybrid TPC/TSC welding” are required. Van Toren proposes in a patent application [8] two approaches to ensure that a thermoset composite coated with a thermoplastic-rich layer does not reach its maximum operating temperature during high-temperature welding. One of them is based on the usage of heat sinks either within the thermoplastic coating or externally applied to it. The other one relies on, as quoted from [8], “the thermoplastic coating having a heat capacity per unit length larger than the heat applied to the assembly per unit length”. Preferred welding processes proposed in this patent application are induction, resistance and laser welding. No experimental results to support these approaches to prevent degradation are, however, presented in this document. Ageorges et al. also showed in [9] that, by using a glass-fibre reinforced hybrid coating layer co-cured with the thermoset composite adherend, resistance welds between CF (carbon fibre)/PEI and CF/epoxy (+hybrid coating layer) composites with acceptable strength levels could be obtained. Even though thermal degradation of the epoxy resin was not experimentally assessed in that paper, the acceptable strength levels obtained might indicate however that no significant degradation took place. From our viewpoint, the usage of glass, and hence thermally insulating, fibres in the thermoplastic coating of the CF/epoxy adherends contributed to shielding the epoxy resin from the relatively high temperatures developed during welding. However, this method introduces a foreign material at the welding interface, i.e. glass fibre fabric, which typically has a lower strength to failure than the adherends themselves.

Our alternative novel approach to preventing thermal degradation of the thermoset resin during high-temperature hybrid TPC/TSC welding is based on considerably reducing the welding time. Very short heating times during the welding process result in: (a) a very short time for heat to be transferred through the thermoplastic coating layer used to generate adhesion with the thermoset composite adherend, which will decrease the temperature at which the thermoset composite adherend is exposed to during the welding process; and (b) a very short time for degradation mechanisms to occur in the thermoset composite adherend, which if short enough, could potentially hinder the occurrence of any degradation [10]. Very short heating times during the welding process are enabled through the usage of ultrasonic welding, which features heating times in the order of fractions of a second as opposed to, for instance, resistance or induction welding, with heating times in the order of minutes.

This paper presents the results of a fully experimental study focused on analysing whether thermal degradation can be prevented during welding of CF/PEEK to CF/epoxy composites through the use of ultrasonic welding and very short heating times. It must be noted that this is a particularly challenging material combination owing to the ample gap between the melting temperature of PEEK and the glass transition temperature of epoxy. Two sets of welding parameters with significantly different average heating times, namely 460 and 830 ms, were used in order to assess the effect of heating time on degradation during ultrasonic welding of CF/PEEK onto CF/epoxy through a co-cured neat PEEK coating layer, referred to as indirect welding hereafter. Direct ultrasonic welding of CF/PEEK onto CF/epoxy, i.e. with no PEEK coating on top of the CF/epoxy adherend, was as well performed in order to investigate the sole effect of short heating times on hindering the degradation reactions in the epoxy resin. Due to the difficulties to measure temperatures during ultrasonic welding without disturbing the heat generation process, degradation assessment was based on the analysis of the fracture surfaces of the CF/PEEK to CF/epoxy welded joints.

2 EXPERIMENTAL

2.1 Materials and sample preparation

The materials used in this study were five-harness satin CF/PEEK from Ten Cate Advanced Composites, The Netherlands, and Hexply 913 unidirectional CF/epoxy from Hexcel. Stacks of powder-impregnated CF/PEEK with a $[0/90]_{3S}$ stacking sequence were consolidated in a hot-platen press at 385°C and 10 bar for 20 min. The CF/PEEK consolidated laminates had a nominal thickness of 1.92 mm. The unidirectional CF/epoxy tapes were automatically laid on a $[0]_{11}$ configuration. According to the specifications of the manufacturer, the CF/epoxy laminates were cured in an autoclave at 125°C and 6 bar for 60 min. A stainless steel caul plate was used to ensure a flat surface on the side of the vacuum bag. The final thickness of the CF/epoxy laminates was 1.7 mm. The glass transition temperature of the CF/epoxy laminates was 157°C, as measured via differential scanning calorimetry (Sapphire DSC Perkin Elmer).

A 0.25 mm-thick neat PEEK film (Victrex, UK) was co-cured to one of the sides of some of these CF/epoxy laminates, referred to as “PEEK-coated CF/epoxy” hereafter. The surface of the PEEK film directly in contact with the CF/epoxy prepreg was degreased and subsequently subjected to ultraviolet ozone cleaning prior to the laying-up process.

CF/PEEK and CF/epoxy adherends, 25.4 mm-wide and 101.6 mm-long, were water-jet cut out of the composite laminates. The adherends were cut so that their longer side coincided with the main apparent orientation of the fibres in the CF/PEEK laminates and with the 0° orientation of the fibres in the CF/epoxy laminates.

2.2 Welding

The two types of welded joints shown in Figure 1 were considered in this study. One of them was a weld between a CF/PEEK adherend and an uncoated CF/epoxy adherend, referred to as “direct weld”. The other one was a weld between a CF/PEEK adherend and a PEEK-coated CF/epoxy adherend, referred to as “indirect weld”. In both cases 0.25 mm-thick flat energy directors were used. A flat energy director is a neat layer of matrix resin, PEEK in this case, that is placed at the welding interface prior to the welding process. It concentrates heat generation at the welding interface through combined surface friction and viscoelastic friction and provides 100% welded areas [11].

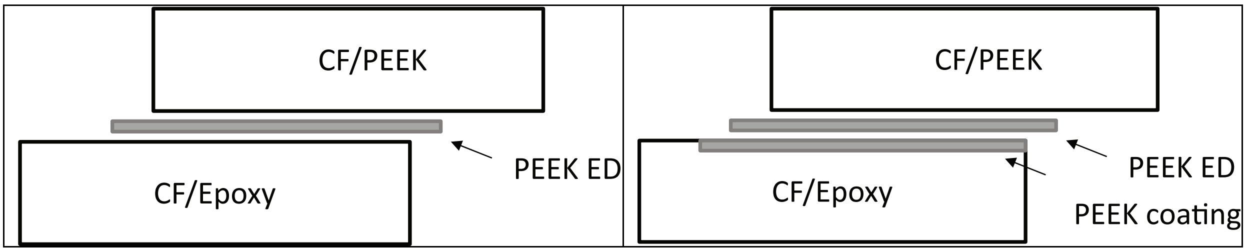

Figure 1. Different types of welded joints considered in this paper. Left: direct weld (CF/PEEK to uncoated CF/epoxy). Right: indirect weld (CF/PEEK to PEEK-coated CF/epoxy). A flat PEEK energy director (PEEK ED) is used in both cases. Dimensions are not to scale.

The CF/PEEK and (coated or un-coated) CF/epoxy adherends were welded in a single lap shear configuration with a 12.7 mm long overlap. A Rinco Dynamic 3000 microprocessor-controlled ultrasonic welder was used for this purpose. An in-house designed welding jig was used to ensure no shifting of the samples and to allow vertical movement, and hence no bending, of the top adherend during the welding process [12]. A 30 mm x 15 mm rectangular sonotrode was used. Adherends and flat energy directors were degreased prior to the welding process.

Two edge combinations of welding force and amplitude, namely high force (1500 N, i.e. 4.7 MPa welding pressure) / high amplitude (90 µm peak to peak) and low force (300 N, i.e. 0.9 MPa welding pressure) / low amplitude (60 µm peak to peak) were used in order to work with the two different heating times, i.e. "short heating time" and "long heating time" shown in Table 1. Displacement-controlled welding was used in every case and optimum displacement values for each force/amplitude combination were determined through the power and displacement data provided by the ultrasonic welder following the procedure explained in [13]. This procedure allows defining an optimum displacement value that ensures that melting of the resin in the first layer of the thermoplastic composite adherends occurs during the welding process without excessive bulk heating. Melting of the resin in the first layer of the thermoplastic composite adherend occurs once the flat energy director is fully melted and flows under the effect of the welding force [14]. Changing of the welding force and the vibration amplitude causes changes in the heat generation rate at the welding interface and hence in the time needed to reach the optimum displacement [14]. Changes in the maximum temperature reached at the welding interface, which is difficult to measure without affecting the welding process, could as well be expected. At least five samples were welded per joint type (see Figure 1) and set of welding parameters.

Reference	Welding force (N)	Peak-to-peak amplitude (µm)	Optimum displacement (mm)	Average heating time (ms)
Short heating time	1500	90	0.10	460
Long heating time	300	60	0.08	830

Table 1: Summary of welding conditions for both direct and indirect welds.

2.3 Analysis and testing

Welded samples were either mechanically tested following ASTM D 1002 standard in a Zwick 250 KN universal testing machine (1.3 mm/min cross-head speed) or manually broken open when adhesion was poor. Degradation on the welding surface of the CF/epoxy adherend was assessed as a first approximation through fractography. No temperature measurements were performed in this study since previous research proved it difficult to place thermocouples at the welding interface without significantly affecting heat generation during the ultrasonic welding process. However, due to the way in which the process parameters were defined, it could be assumed that the maximum temperature reached at the welding interface was, in any case, equal or above the melting temperature of PEEK.

3 RESULTS AND DISCUSSION

3.1 Direct welds

Welding of CF/PEEK adherends directly onto CF/epoxy adherends provided samples with a low degree of adhesion that could be separated manually. For the long-welding-time conditions (Table 1), a relatively big area of apparently dry fibres was observed on the fracture surfaces of the CF/epoxy adherends (see Figure 2). Detailed inspection of this area under the scanning electron microscope (SEM) revealed lack of resin connecting the fibres, which became more accentuated towards one of the edges of the overlap. The fracture surfaces of the CF/epoxy adherends for the short-welding-time conditions (Table 1) showed, however, a much more contained footprint of the welding process on the CF/epoxy adherends (Figure 3). Seemingly dry fibres with a similar aspect to the ones in Figure 2 could be observed on one of the edges of the overlap as well as some detached fibre bundles at the edges of the sample and some longitudinal cracks. However, a big fraction of the fracture surface did not show visible differences with the material outside the welding area. The non-uniform appearance of the fracture surfaces in Figures 2 and 3 seems to be consistent with the fact that, during the welding process, heat is generated at a faster rate at the edges of the welding area [15]. The fact that one of the edges of the overlap seemed less affected than the rest is consistent with the existence of differences in heat transfer between both overlap edges caused by the asymmetric design of the welding jig used in this research [14].

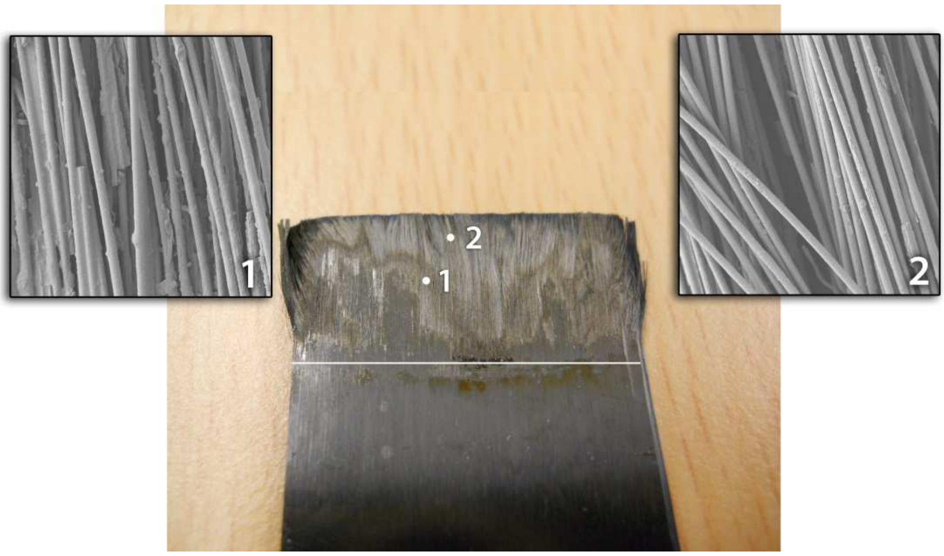

Figure 2: Fracture surface of the CF/epoxy adherend after direct welding under long-welding-time conditions (830 ms heating time) and SEM details. 1: Some resin is still present on and between the fibres. 2: Absence of resin connecting the fibres towards the edge of the overlap. White line delimits welding overlap

These results suggest that one of the ways thermal degradation manifests itself on the welding surface of the CF/epoxy is through the presence of dry carbon fibres, probably due to the vaporisation of the epoxy resin owing to the high temperatures reached during the welding process. According to the results of thermo-gravimetric analysis of the CF/epoxy composite used in this study, a 25% weight loss can be expected at 500°C (Pyris Diamond TGA, Perkin Elmer, 20°C/min heating rate), which may indicate welding temperatures well above 500°C towards the hottest overlap edge. Nevertheless, the effect of reducing the heating time from 830 to 460 ms can be clearly observed on the welding surface of the CF/epoxy adherends. Even though lowering the heating time to 460 ms does not fully prevent the presence of dry carbon fibres, and supposedly thermal degradation, at the hottest edge of the overlap, it does significantly reduce the impact of the welding process on the CF/epoxy adherend.

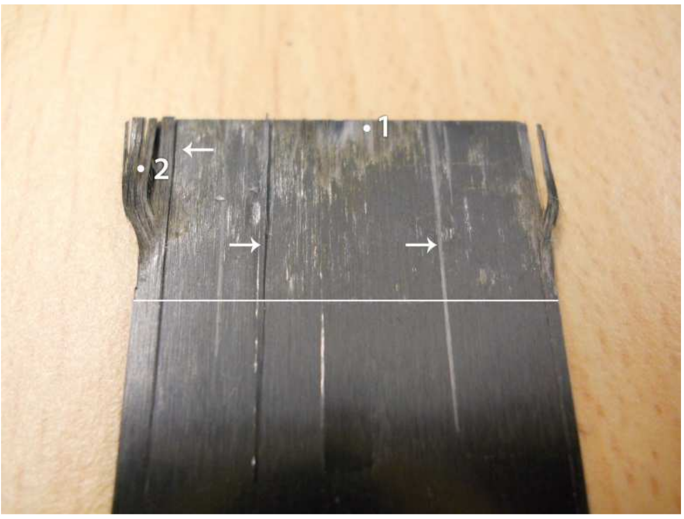

Figure 3: Fracture surface of the CF/epoxy adherend after direct welding under short-welding-time conditions (460 ms heating time). 1: Dry fibres. 2: Detached fibre bundles. Arrows indicate position of longitudinal cracks. White line delimits welding overlap.

3.2 Indirect welds

Welding of CF/PEEK adherends onto CF/epoxy adherends through a co-cured PEEK coating layer resulted in stronger welds that had to be pulled apart through mechanical testing in order to access the fracture surfaces. Samples welded under long-welding-time conditions (Table 1) showed a combination of two failure types: (i) adhesive failure between the co-cured PEEK layer and the CF/epoxy adherend, and (ii) failure in the CF/epoxy adherend (see Figure 4). As also shown in Figure 4, detailed SEM analysis of the CF/epoxy failure revealed dry and disconnected fibres, such as the ones found on the thermally degraded surfaces in Figure 2, at the very edge of the hottest side of the overlap. Nevertheless, most of the CF/epoxy failure area showed resin-rich surfaces with no sign of thermal degradation in the form of rupture or absence of the resin in between the fibres. The samples welded under short-welding-time conditions (Table 1) showed only adhesive failure between the co-cured PEEK layer and the CF/epoxy adherend and no visible surface damage as seen in Figure 5.

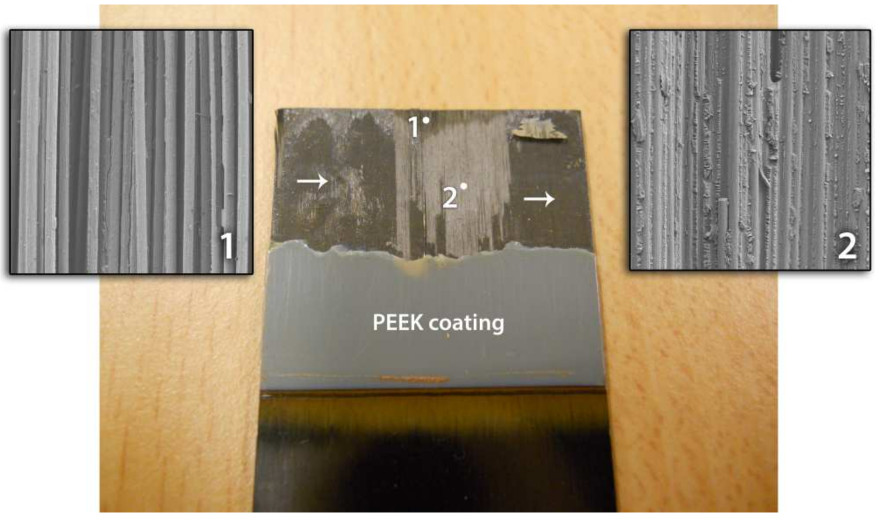

Figure 4: Fracture surface of the CF/epoxy adherend after indirect welding under long-welding-time conditions (830 ms) and SEM details. 1: CF/epoxy failure at the very edge of the overlap where thermal degradation is suspected. 2: CF/epoxy failure in the rest of the overlap featuring a resin-rich fracture with no visible signs of thermal degradation. Arrows indicate areas with adhesive failure.

According to these results, the 0.25 mm-thick PEEK coating layer succeeded in preventing observable thermal degradation on the CF/epoxy adherends when the short-heating-time conditions were used. This is explained by the fact that, by using the PEEK coating layer, the location at which heat generation occurs during the welding process is moved away from the surface of the CF/epoxy adherend to the interface between the PEEK coating and the energy director. Consequently, the temperature at which the surface of the CF/epoxy substrate was exposed to during the welding process depended on the heat conducted through the PEEK coating layer during a very limited amount of time and, hence, could be expected to be significantly lower than the temperature at the welding interface.

Figure 5: Fracture surface of the CF/epoxy adherend after indirect welding on a co-cured PEEK layer under short-welding-time conditions (460 ms). Only adhesive failure can be observed here.

The importance of short heating times to prevent degradation was demonstrated by the fact that for the long-heating-time conditions some initial and very local degradation could already be observed at the hottest edge of the overlap in the form of a small area of dry and disconnected fibres on the CF/epoxy substrate. Given the significant gap in terms of force and amplitude between the two sets of welding parameters considered in this study (see Table 1), these results indicate the potential existence of a wide range of combinations of processing parameters providing welds without thermal degradation of the CF/epoxy adherend for this particular material combination.

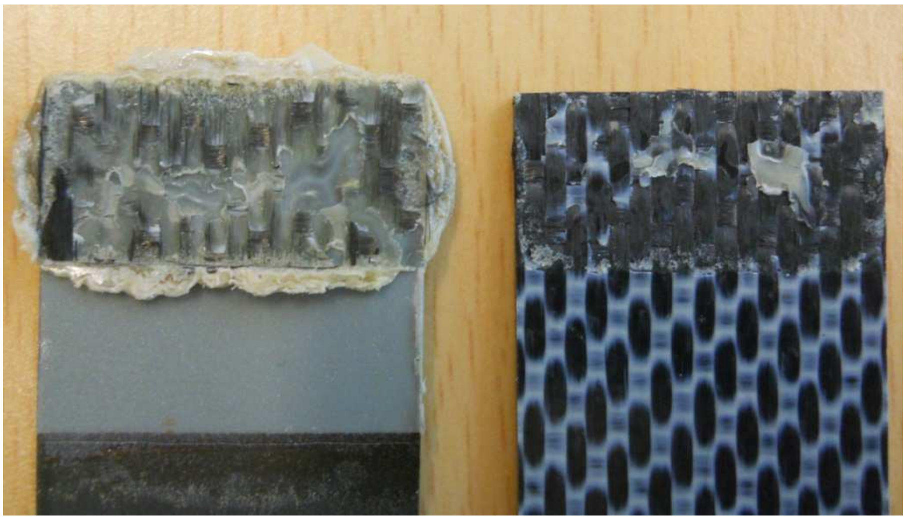

Figure 6. Fracture surfaces of a CF/PEEK to CF/epoxy indirect weld with improved adhesion between the CF/epoxy and the PEEK coating layer co-cured to it. Failure mostly occurs in the CF/PEEK composite substrate (right).

As a step further in this research, an improved co-curing procedure was used to obtain PEEK-coated CF/epoxy adherends with higher adherence between the 0.25 mm-thick PEEK coating and the CF/epoxy material. These improved adherends were welded to CF/PEEK samples under the short-heating-time conditions (Table 1) and mechanically tested afterwards. A relatively high lap shear strength amounting to 29.6±1.9MPa (average and standard deviation values from ten samples) was obtained. But more importantly, the fracture surfaces showed that failure primarily occurred at the CF/PEEK substrate (see Figure 6), confirming the lack of any significant damage in the CF/epoxy substrate during the welding process.

4 CONCLUSIONS

A fully experimental study focused on prevention of degradation during high-temperature welding of CF/PEEK (Tm=340°C) and CF/epoxy (Tg=157°C) composites was presented in this paper. A novel approach to prevent degradation of the CF/epoxy composite during the welding process based on the usage of very short welding times was followed. Fractographic analysis of welded and subsequently tested samples was carried out in order to assess the occurrence of thermal degradation of the CF/epoxy composite.

The most important conclusion of this work is that high-temperature hybrid TPC/TSC welding without detrimental thermal effects on the TSC adherend can be achieved with the combined effect of a thin thermoplastic coating layer on the TSC adherend, which acts as an effective heat shield, and heating times for the welding process well under 1 second, which are only offered by ultra-fast welding techniques such as ultrasonic welding.

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REFERENCES

[1] A. Offringa, Thermoplastics in aerospace, a stepping stone approach. *CETEX thermoplastic composites: from scratch to flight*, Delft University of Technology, Delft, 2006.

[2] A. Deters, A. Miaris, G. Sohnes, Serial production of thermoplastic CFRP parts for the Airbus A350XB. *Proceedings of the 1st ITECH Conference*, Bremen, October 2012.

[3] C. Ageorges, L. Ye, M. Hou, Advances in fusion bonding techniques for joining thermoplastic matrix composites: a review. *Composites: Part A*, **32**, 2001, pp. 839-857.

[4] R.C. Don, J.W. Gillespie Jr., S. H. McKnight, Bonding techniques for high performance thermoplastic compositions. *United States Patent No. 5643390*, 1997.

[5] M. Hou, A. Beehag, Q. Yuan, Welding techniques for polymer or polymer composite components, *International patent No. 011573*, 2003.

[6] G.J. Jacaruso, G. C. Davis, A. J. McIntire, Method of making thermoplastic adhesive strip for bonding thermoset composite structures, *United States Patent No. 5264059*, 1993.

[7] M. Hou, Thermoplastic adhesive for thermosetting components, *Materials Science Forums*, **706-709**, 2012, pp 2963-2973.

[8] M. Van Toren, Method for bonding a thermoplastic polymer to a thermosetting polymer component, *International Patent Application No. 161569A1*, 2012.

[9] C. Ageorges, L. Ye, Resistance welding of thermosetting composite/thermoplastic composite joints, Composites: Part A, **32**, 2001, pp. 1603-1612.

[10] E.D. Wetzel, R.C. Ron, J.W. Gillespie Jr., Modelling thermal degradation during thermoplastic fusion bonding of thermoset composites, *Proceedings of the 52nd Annual Technical Conference, ANTEC'94*, 1994.

[11] I.F. Villegas, B. Valle Grande, H.E.N. Bersee, R. Benedictus, A comparative evaluation between flat and traditional energy directors for ultrasonic welding of thermoplastic composites, *Proceedings of the 16th European Conference on Composite Materials, ECC16*, Seville, Spain, 2014.

[12] I.F. Villegas, H.E.N. Bersee, Ultrasonic welding of advanced thermoplastic composites: an investigation on energy-directing surfaces. *Advances in Polymer Technology*, **29**(2), 2010, pp. 112–21.

[13] 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*, **65**, 2014, pp. 27-37.

[14] I.F. Villegas, In-situ monitoring of ultrasonic welding of thermoplastic composites through power and displacement data, *Journal of Thermoplastic Composite Materials*, **28**(1), 2015, pp. 66-85.

[15] A. Levy, S. Le Corre, I.F. Villegas, Modelling of the heating phenomena in ultrasonic welding of thermoplastic composites with flat energy directors, *Journal of Materials Processing Technology*,**214**, 2014, pp. 1361-1371.