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ULTRASONIC WELDING OF CF/EPOXY TO CF/PEEK COMPOSITES: EFFECT OF THE ENERGY DIRECTOR MATERIAL ON THE WELDING PROCESS

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Abstract

With its short heating times, ultrasonic welding is a highly promising technique for joining thermoplastic (TPC) to thermoset (TSC) composites, to prevent thermal degradation of the thermoset adherend. A neat thermoplastic coupling layer is co-cured on the surface to be welded to make the TSC "weldable". For welding CF/PEEK to a TSC adherend, it would be logical to use PEEK as the coupling layer. However PEEK and epoxy are not miscible with each other, therefore a bond created after co-curing of these two materials is not reliable. PEI on the other hand is known to be miscible to most epoxy systems at high temperatures and PEEK polymers, hence it is an excellent candidate for the coupling layer material. The other necessary element for ultrasonic welding is the energy director (ED), a neat TP film placed at the interface to help promote heat generation through preferential frictional and viscoelastic heating. Usually EDs are made from the same material as the TP matrix, but in this case ED can be either PEI or PEEK. Mechanical testing and fractographic analysis showed that the usage of a PEEK ED is the most successful approach. This research is part of the European project EFFICOMP.

1. Introduction

Ultrasonic welding is an excellent technique for bonding composites as it can provide strong joints in a rather fast and cost- effective in terms of manufacturing way [1, 2]. Its exceptionally short heating times of less than 1 sec is what makes ultrasonic welding the most competent welding method for joining TPC to TSC because, as shown in the research of Villegas and Rubio[3], it can prevent thermal degradation of the TSC adherend. Despite the high potential of the ultrasonic welding process, not much research has be done on the usage of the technique for hybrid composite welding. Lionetto et al [4] welded two CF/epoxy adherends through PVB films using both induction and ultrasonic welding and demonstrated good results for both cases. Villegas and van Moorleghem also presented good results for ultrasonic welding of CF/epoxy to CF/PEEK composites through a PEI coupling layer. Most of the study was focused on the epoxy/ PEI interphase formation and only mechanical testing results were presented with no fractographic analysis[5]. In a previous work the authors of this paper investigated the possibility of welding CF/epoxy to CF/PEI without a loose ED, solely through the coupling layer, and concluded that for welding of advanced composites an ED is always required at the interface to help promote heat locally, without risking excessive bulk heating. The good results obtained in all the mentioned studies show how promising ultrasonic welding is for hybrid joints[6]. Still, the full potential of this welding technique is not explored and therefore this study aims at adding more knowledge on its possibilities and limitations.

The main focus of this study is to explore different approaches for welding a TSC containg a TP coupling layer to a TPC whose matrix is a different material from the coupling layer. The existence of two TP materials at the welding stack gives rise to the question of what the material of the ED should be in order to produce a high-performance weld. Mismatch of the melting temperatures and thermal properties of the two TP materials may have a significant effect on the welds. Therefore, the present paper aims at assessing what the effect of the nature of the ED material on the mechanical performance of the welds is by performing single lap shear tests, followed by microscopic investigation to fully understand the effect of the different welding stacks.

2. Experimental Procedure

2.1 Materials

In this study, Cetex® CF/PEEK (carbon fibre/polyetherimide) with a 5-harness satin fabric reinforcement, manufactured by TenCate Advanced Composites, and T800S/3911 unidirectional CF/epoxy from TORAY, were used. CF/PEEK preimpregnated laminates with a $[0/90]_{3s}$ stacking sequence were consolidated in a hot-platen press at 385 °C and 1 MPa for 20 min. The thickness of the consolidated laminates was around 2 mm. Unidirectional CF/epoxy pre-preg was manually laid up in a $[[0/90/0/90]_s$ configuration. Note that the CF/epoxy pre-preg contained TP toughening particles at both surfaces the nature of which not provided by the manufacturer. A 0.06mm-thick neat PEI film (SABIC), was used as the coupling layer and it was co-cured to one of the sides of the CF/epoxy laminates. The PEI coupling layer was degreased with isopropanol prior to its application on top of the pre-preg stack. The CF/epoxy laminates with the coupling layer were cured in an autoclave at 180°C and 7 bars for 120 min, according to the specifications of the manufacturer. To ensure flat surfaces on both sides of the laminate, an aluminium caul plate was used on the side of the vacuum bag. The final thickness of the CF/epoxy/PEI laminates was also approximately 2 mm. A gradient interphase was formed between the epoxy and PEI material after curing with a thickness of approximately 25 μm. The interphase consisted of epoxy spheres that were dispersed into the PEI with a size that was decreasing as the PEI content was increasing. More information on how the interphase was developed can be found in the authors' previous work in reference [6].

CF/PEEK and CF/epoxy/PEI adherends with dimensions 25.4mm x 101.6mm were cut from the laminates using a water-cooled circular diamond saw. The CF/PEEK adherends were cut with their longitudinal direction parallel to the main apparent orientation of the fibres. The CF/epoxy/PEI adherends were cut with their longitudinal direction parallel to the 0 fibres.

2.2 Welding process

To assess the effect of the nature of the ED material on the mechanical performance of the welds, four welding configurations were considered which are presented in Table 1. Individual samples were welded with a Rinco Dynamic 3000 ultrasonic welder in a single lap configuration, with the overlap being 12.7 mm long and 25.4 mm wide, using the custom-made setup, shown in Fig 1. A cylindrical sonotrode with a 40 mm diameter was utilised. To ensure minimum heating times, and hence minimum risk of thermal degradation at an acceptable level of dissipated power, the parameters chosen were 1500 N welding force and 86.2 μm peak-to-peak vibration amplitude [3]. Solidification force and time were kept constant at 1500 N and 4 s respectively. Displacement-controlled welding was used, in which the vibration time was indirectly controlled by the displacement of the sonotrode, as it was shown to provide high-strength welds with minimum scatter in previous research [7]. The optimum welding displacement was obtained from the feedback data provided by the ultrasonic welder following the procedure defined in reference [8].

Table 1. The different welding approaches investigated in this paper.

Welding case	Referred to as:
CF/epoxy/PEI welded to CF/PEEK through a PEEK ED	Hybrid PEEK
CF/epoxy/PEI welded to CF/PEEK through a PEI ED	Hybrid PEI
CF/PEEK welded to CF/PEEK through a PEEK ED	Reference PEEK
CF/PEEK welded to CF/PEEK through a PEI ED	Reference PEI

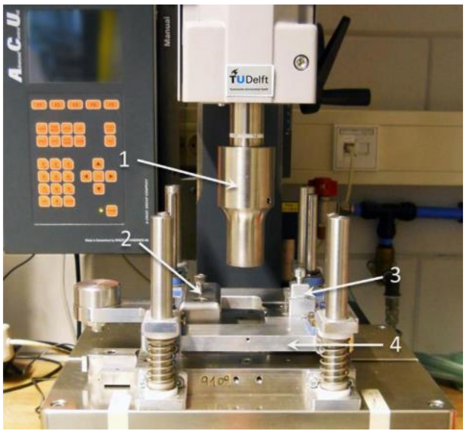


Figure 1. Custom made welding setup. 1: sonotrode, 2: clamp for the lower sample, 3:clamp for the upper sample and 4: sliding platform [9].

2.3 Testing

The mechanical performance of the welded samples was assessed by performing single lap shear tests based on the ASTM D 1002 standard in a Zwick 250 kN universal testing machine. The apparent lap shear strength (LSS) of the joints was calculated as the maximum load measured during testing divided by the overlap area. Five specimens were welded per welding case to determine the average LSS. Naked eye observation, and scanning electron microscopy (SEM, JEOL JSM-7500F scanning electron microscope) were used for the fractographic analysis of the welded joints.

3. Results

Fig. 2 illustrates the results obtained from the single lap shear mechanical testing. Both hybrid configurations resulted in a similar LSS, 34.9 ± 1.4 MPa and 31.8 ± 2.9 MPa for the hybrid PEEK and hybrid PEI respectively. Hybrid PEI presented a slightly bigger scatter. Reference PEEK samples yielded the highest LSS, i.e. 44.8 ± 4.4 MPa (almost 28% higher than the corresponding hybrid case) but also the highest scatter. Reference PEI specimens yielded a 37.8 ± 1.8 MPa LSS which is 16% lower as compared to the reference PEEK samples and 18% higher than the corresponding hybrid case.

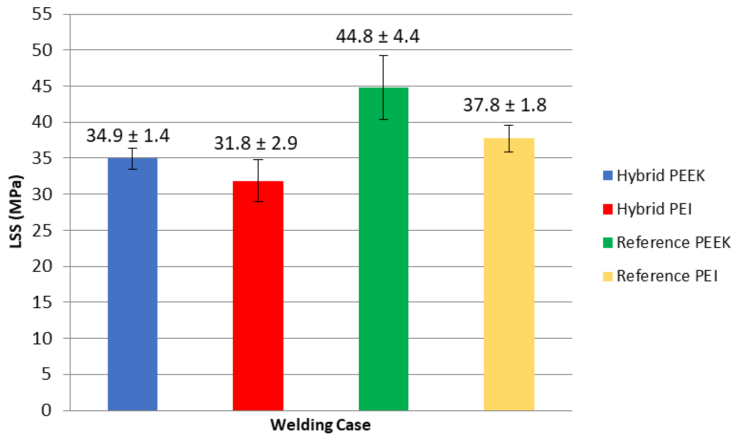
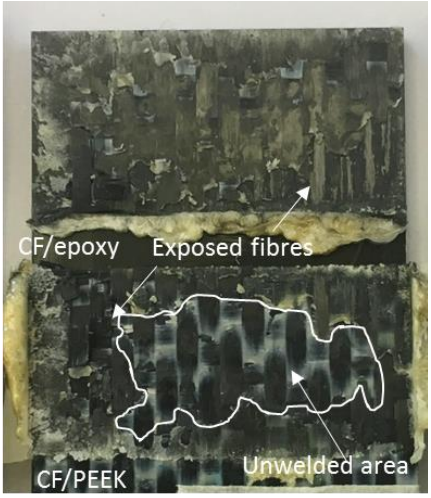


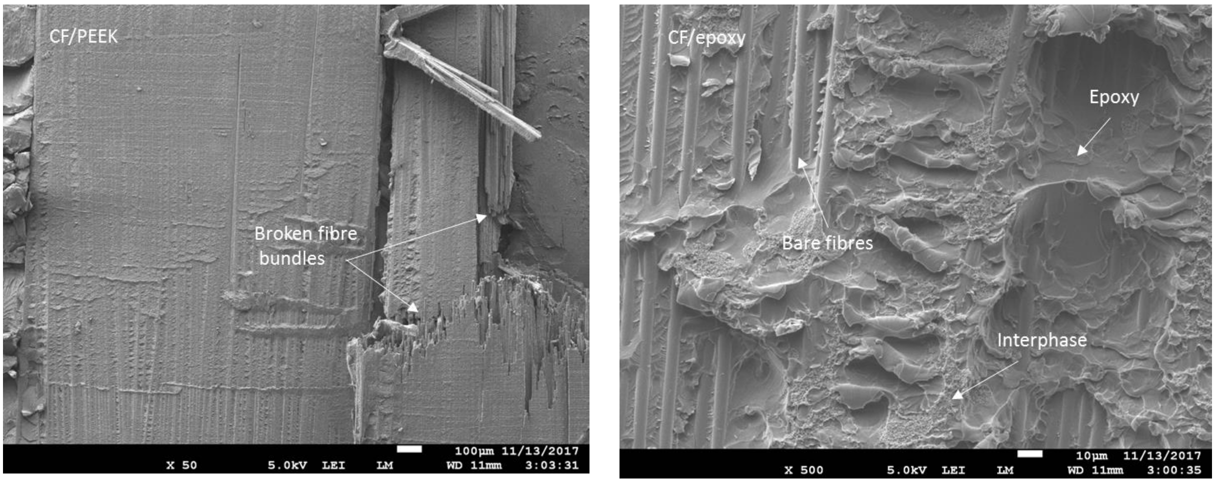
Figure 2. LSS values for the different welding configurations

Fig. 3a and Fig. 3b depict representative fracture surfaces of a hybrid PEEK sample and a hybrid PEI sample respectively. The top fracture surface corresponds to the CF/epoxy adherend and the bottom to the CF/PEEK adherend of the hybrid weld. Unwelded areas were present in the centre of the overlap of both samples. For the hybrid PEEK configuration (Fig. 3a) they covered around 20% of the total overlap and for the hybrid PEI samples (Fig. 3b) they covered approximately 50% of the overlap. Failure for both configurations seemed to occur in both composite adherends, as fibres and resin from one composite substrate could be found on the other substrate. Fig. 3c is a SEM image showing failure in the CF/PEEK, which corresponds to the circled area (c) on Fig. 3a. It shows fibre bundles from the CF/PEEK adherend that are fully covered with resin and some of the fibre bundles are broken. Closer inspection of the circled area (d) in Fig. 3a reveals that the failure in the CF/epoxy adherend was characterized by bare fibres, failure in the epoxy matrix and interphase failure, since the epoxy spheres can be seen on the fracture surface (Fig. 3d). The failure mechanisms analyzed above were also observed for the hybrid PEI configuration, hence the microscopic analysis reported in this section also applies to the latter case.

a



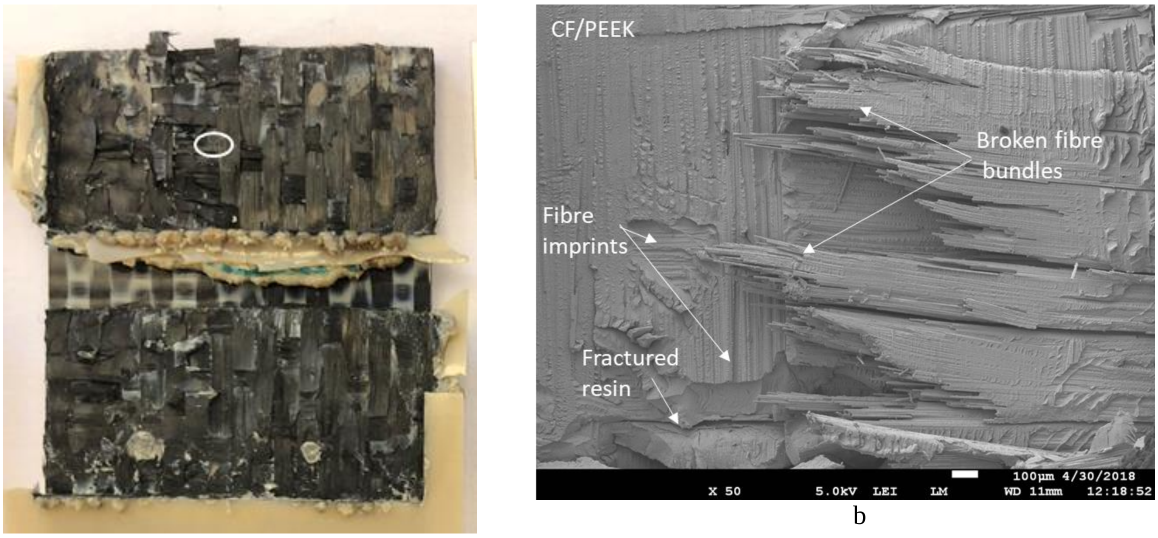
b



c d

Figure 3. Representative fracture surfaces of a hybrid PEEK sample (a) and a hybrid PEI sample (b), showing unwelded areas and failure in CF/PEEK and CF/epoxy adherends, c) and d) are SEM images of the circled areas in a) showing failure in the CF/PEEK and failure in the CF/epoxy respectively.

Fig. 4a shows the representative fracture surfaces of a reference PEEK sample. Fully welded areas and broken fibre bundles are observed, some of which most likely come from the second ply of the adherend. Fig. 4b is a SEM image of the circled area in Fig. 4a, illustrating broken fibres, fibre imprints on the matrix and fractured resin.

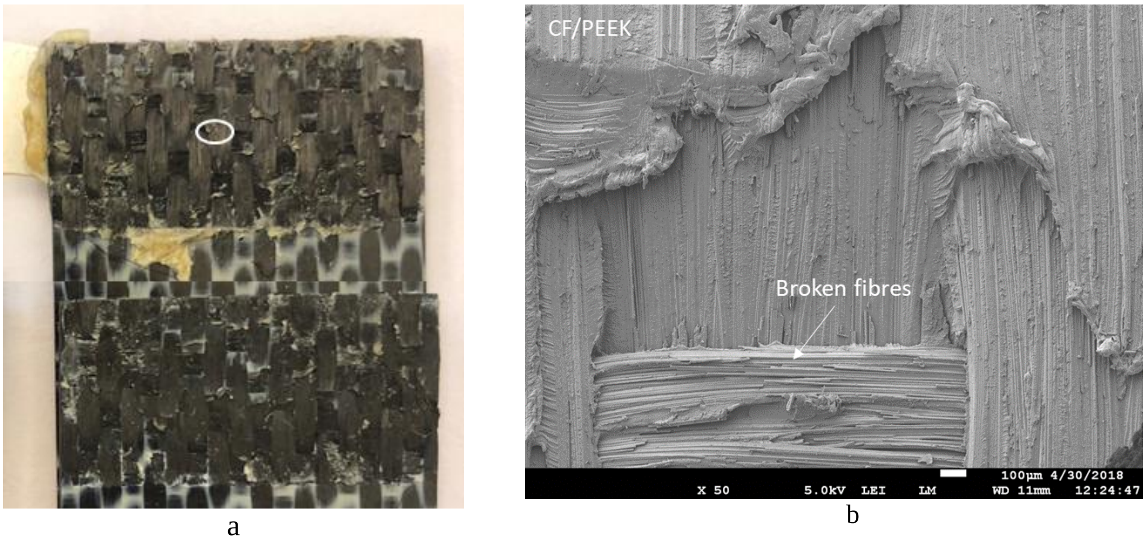


b

a

Figure 4. a) Representative fracture surfaces of a Reference PEEK sample showing failure deep into the composite and b) is a detailed SEM image of the circled area in a) showing broken fibres, fibre imprints and broken resin.

Lastly, Fig. 5a presents the fracture surfaces of a reference PEI sample which shows fully welded areas. The surfaces appear to be smooth with no broken fibre bundles being visible with naked eye. The SEM image of Fig. 5b shows fibre bundles that are fully covered with resin for the most part and some broken fibres.



a b

Figure 5. a) Representative fracture surfaces of a Reference PEI sample showing exposed fibres and b) is a SEM image of the circled area in a) showing broken fibres and resin rich bundles.

Discussion

The different LSS values obtained from mechanical testing (Fig.1) can be explained by comparing the failure modes of each welding configuration. First of all, comparing the two hybrid cases reveals unwelded areas the amount of which is different for each configuration. Unwelded areas seem to always occur for hybrid welds in which the coupling layer is a thin PEI film and the TPC is CF/PEEK [5], but not when the TPC is CF/PEI [6]. A possible explanation for these unwelded areas might be the mismatch of the temperatures at which the PEEK and PEI resins flow. In the hybrid PEI configuration the PEI ED and coupling layer start flowing and being squeezed out the weld zone at a temperature above the T_g (217 ^oC) which is well below the melting temperature of the PEEK matrix (343 ^oC). Thus, areas of the CF/PEEK adherend are left unmolten, preventing the PEEK matrix to blend with the PEI ED and vice versa. Using a PEEK ED instead helps decrease the unwelded areas since the PEEK ED starts melting at the same temperature as the PEEK matrix. However, the fact that the unwelded areas are not completely eliminated might be attributed to earlier melting of the PEI coupling layer as compared to the PEEK matrix. Melting and flow of the PEI coupling layer together with the PEEK ED might lead to reaching the targeted displacement before full melting of the PEEK resin on the CF/PEEK surface was achieved. Welding with a higher displacement might help eliminate the unwelded areas but also increase the risk of thermal degradation of the CF/epoxy adherend.

The mechanical testing results reported in Fig.1 show that the nature of the material of the ED does not have a significant impact on the average LSS of the hybrid joints, even though the two hybrid configurations have quite different amount of unwelded areas. A possible explanation as to why the unwelded areas do not seem to affect the strength of the welds considerably can be the fact that the unwelded areas are concentrated in the middle of the overlap. The edges, where the high stresses are developed, are in both cases welded and show similar failure in both composite substrates, as seen in Fig. 3a and 4a. The higher scatter for the hybrid PEI configuration is attributed to the different amount of welded areas of every sample. Therefore, the hybrid PEEK samples seemed to have a better mechanical performance since they consistently yielded a higher LSS and bigger welded areas.

Secondly, comparison between the hybrid PEEK and reference PEEK configurations reveals a considerably higher LSS for the latter. The reference PEEK configuration resulted in fully welded areas (Fig. 4a) and the post testing failure was characterized by broken fibre bundles, fibre imprints and matrix failure (Fig. 4b) in the first ply (locally even second ply), which combined with the high

LSS, indicate good mechanical performance of the reference PEEK samples. On the other hand the hybrid PEEK samples showed unwelded areas and first ply failure in both CF/PEEK and CF/epoxy adherends. Failure in the latter was characterized also by epoxy and interphase failure. Failure in the PEEK resin in the reference samples versus the epoxy failure in the hybrid PEEK samples might result in a decreased LSS since the PEEK matrix could be expected to have a higher toughness than the epoxy resin. The higher scatter in the reference PEEK configuration is most likely attributed to how deep was the failure in the CF/PEEK, which varied per sample.

Lastly, both reference configurations resulted in fully welded areas, however the Reference PEI fracture surfaces were flat and much smoother than the Reference PEEK (Fig. 6a). As seen in Fig. 6b the failure in the composite was not as deep as in the reference PEEK samples and less broken fibres could be found on the overlap. The deep composite failure is probably a sign of high crack propagation resistance in the adherend which can result in a high joint strength. Thus, the reference PEI samples yielded a 16% lower LSS as compared to reference PEEK samples. Note that the strength of the hybrid samples is comparable to the strength of the reference PEI samples showing once more the promising results of the latter configuration.

The differences in welded areas and also failure modes of the different welding configurations might be linked to the heat generation, i.e. frictional and viscoelastic heating and the heat transfer, i.e. heat capacity and thermal conductivity of the materials involved in each case. Further research on the material properties associated to the mentioned mechanisms will allow for a complete understanding of the effect of the nature of the ED material on the welding process and weld quality.

Conclusions

In this paper, experimental assessment of the effect of the nature of the ED material on the mechanical performance on ultrasonically welded joints was presented. Four different welding configurations were considered, 1) two hybrid, i.e. one CF/epoxy and one CF/PEEK adherend and 2) two CF/PEEK adherends, each configuration utilizing both PEEK and PEI ED. Analysing the results presented in the previous section led to the following conclusions:

- For hybrid welds, using a PEEK ED resulted in unwelded areas that covered around 20% of the total overlap and samples yielded an average LSS of 34.9 ± 1.4 MPa. Using a PEI ED resulted in bigger un-welded areas which were around 50 % of the total overlap and a LSS with a higher scatter, i.e. 31.8 ± 2.9 MPa. Broken samples from both configurations showed similar type of failure in both composite adherends.

- The hybrid PEEK samples yielded a 22% lower LSS as compared to the reference PEEK samples. The decrease in the LSS might be attributed to the epoxy failure that occurred in the hybrid samples versus the PEEK failure in the reference PEEK samples. PEEK resin could possibly have a higher toughness as compared to epoxy resin which combined also with the unwelded areas in the hybrid PEEK welds might result in the lower LSS. The hybrid samples however exhibited a relatively close LSS to the one of the reference PEI samples.

Further research on the materials’ thermal and viscoelastic properties can potentially provide a better understanding of the effect of the nature of the ED material on the welding process and weld quality. Nevertheless, the promising results reported in this study demonstrate the high potential of using ultrasonic welding for producing hybrid welds.

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