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Hybrid welding of carbon-fiber reinforced epoxy based composites

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Abstract

The approach for joining thermosetting matrix composites (TSCs) proposed in this study is based on the use of a low melting co-cured thermoplastic film, added as a last ply in the stacking sequence of the composite laminate. During curing, the thermoplastic film partially penetrates in the first layer of the thermosetting composite, leading to macro-mechanical interlocking as the main connection mechanism between the thermoplastic film and the underlying composite. After curing, the thermosetting composite joints with the thermoplastic modified surface can be assembled by welding. Welding of the TSC-TSC joints is performed by ultrasonic and induction welding. The weld strength is investigated by morphological characterization of cross sections and failure surfaces and by mechanical testing. The effect of the thermoplastic film thickness on the welding process and on its outcome is also analyzed. Both induction and ultrasonic welding mostly result in good-quality welded joints. The welding process used as well as the initial thickness of the

thermoplastic film are found to have a significant effect on the final thickness of the weld line and on the location of failure. Thicker thermoplastic films are found to ease the welding processes.

Keywords:

A. Polymer-matrix composites (PMCs);
 B. Adhesion;
 D. Microstructural analysis;
 E. Assembly.

1. Introduction

The competitive advantage of thermosetting matrix composites (TSCs) is offset by the associated assembly costs, which, in the case of aeronautic structures, still roughly represent 50% of the manufacturing costs [1], [2], [3]. Usually, joining of TSCs has been limited to mechanical fastening and adhesive bonding, sometimes a combination of the two. The diffusion of multi-material design requires the development of novel joining techniques able to overcome the drawbacks of mechanical fastening or adhesive bonding [4]. The major drawback of mechanical fastening is the drilling of holes which result in stress concentrations and weakening of the thin composite structure. To reduce the stress concentrations, the number of plies and, thus, the weight of the component is (locally) increased, significantly reducing the lightweight potential of the composite structure. In addition, mechanical fastening is expensive, generally requiring several working hours on drilling equipment, and it adds the extra weight of the fasteners to the composite structure. Adhesive bonding, although not requiring holes, needs both a rigorous surface preparation, high curing temperatures and pressures and long curing times, which are associated to a significant cost increase [5]. Moreover, some factors during manufacturing of structural bonded joints, such as surface treatments, curing cycle of the adhesive or entrapped moisture in the adherends, can strongly affect the long-term durability of bonded composite joints [1], [6].

Lastly, fusion bonding, or welding, is a highly efficient process for joining fiber reinforced thermoplastic composites (TPC), being capable of producing joints in relative short cycle times,

which are characterized by equivalent or better performance than adhesively bonded or mechanically fastened joints [7]. Fusion bonding is based on the melting or softening ability of a thermoplastic polymer with increasing temperature. The associated increase of the mobility of molecular chains enables interdiffusion of the thermoplastic molecular chains of the joint adherends across the joining interface [7]. Several fusion-bonding techniques are available for joining thermoplastic matrix composites, which are based on different heating mechanisms, such as hot plates, hot gas, ultrasonic, microwaves, laser, induction, friction stir, etc. [8], [9], [10], [11]. Fusion bonding is in principle not possible for TSCs since they cannot melt or soften owing to their crosslinked molecular structure. For this reason, the welding of thermosetting composite structures has been scarcely considered in the literature.

In order to enable the use of welding for the joining of thermoset composite structures, Don et al.[12] patented a method for resistance welding of TPCs to cured TSCs based on the co-curing of a thin thermoplastic film of Polyethersulfone (PES) on the thermosetting composite stack. Cocuring of a thermoplastic film, hereafter referred to as "coupling film", with TSCs has been used or suggested for welding of TPCs to TSCs with various coupling layer materials such as Polysulfone (PSU) [13], Polyphenylenesulfide (PPS) [14], Polystyrene (PS), Polyether ether ketone (PEEK) [15] and Polyetherimide (PEI) [16]. In the case of TSC-TSC welded joints, the very scarce information available in the open literature indicates the use of Polyvinylidene fluoride (PVDF) as preferred coupling layer material [5],[17]. Recently, some of the authors of the present paper have used Polyamide 6 (PA6) coupling layers to achieve hybrid joining of aluminum to TSC sheets through ultrasonic metal welding [18].

The selection of the coupling layer material is constrained by various requirements such as processing temperature, high adhesion to the TSC adherend and mechanical and environmental performance of the resulting welded joint. The processing temperature of the coupling layer, i.e. the temperature range at which it can be welded, plays, together with the heating time in the welding process, a crucial role in the potential thermal degradation of the TSC adherend during welding.

Coupling layer materials with welding temperatures lower or similar to the glass transition temperature of the thermosetting resin, such as PVDF, are preferred to minimize the risk of thermal degradation [5],[17]. Coupling layers with higher welding temperatures can as well be used but require additional strategies to prevent thermal degradation and/or deconsolidation, such as an increasing the thickness of the coupling layer [16] or significantly reducing the heating times [15]. Adhesion of the coupling layer material with the TSC adherend affects how strong and durable the connection between the two elements is. A potentially strong and durable connection between the thermoplastic coupling layer and the TSC adherend can be achieved though diffusion of the epoxy monomers into the thermoplastic film during the co-curing process. This will cause local swelling and dissolution of the thermoplastic resulting in an gradient interphase, typically between tenths and hundreds of microns thick, between the pure epoxy and the pure thermoplastic [19], [20]. According to some authors, this process might as well lead to semi-interpenetration of the polymer networks [7]. Nevertheless, the formation of TP-TS gradient interphases and/or semiinterpenetrating networks poses restrictions to the nature of the TP coupling layer material, chiefly related to its solubility in the selected epoxy system. Typically, amorphous thermoplastic polymers, such as polyetherimide (PEI) or polyphenylene ether (PPE), are known to create gradient interphases with some commercial epoxy systems [19]. Amorphous thermoplastic polymers are however known to have low chemical resistance. Alternatively, in the cases where no solubility exists between the coupling layer material and the thermosetting resin in the TSC adherend, connection between the two elements can still be created through macro-mechanical interlocking and/or chemical adhesion. To the end of achieving macro-mechanical interlocking, Jacaruso et al.[21] proposed the use of coupling layers consisting of fabric layers partially impregnated with TP resin. Macro-mechanical interlocking between the coupling layer and the TSC was achieved through flow of the TS resin into the dry fabric areas during the co-curing process. Alternatively, Grefe et al. [22] proposed the use of laser ablation to create texture features on the already cured TSC adherend which led to macro-mechanical interlocking upon melting and flow of the TP resin

into the texture features during the welding process. It should be noted that in this last case no coupling layer was used on the TSC adherend. Finally, attaining chemical adhesion between the thermoplastic coupling layer and the thermosetting resin is a challenging task owing to the chemical inertness of thermoplastic resins. Nevertheless, some promising results have been obtained through UV-O₃ treatment of the TP coupling layer prior to the co-curing process [15].

In this paper, for the first time, a preliminary feasibility study on induction and ultrasonic welding of thermosetting matrix composites, co-cured with a top thermoplastic layer, is presented. Even if the approach of co-curing a thermoplastic film is known for the welding of thermosetting composites, its application to ultrasonic welding and induction welding of thermosetting composites is not present in the literature to the best of our knowledge.

Aiming at widening the currently scarce knowledge on the topic, this paper focused on welding of TSC adherends through a novel low-melting thermoplastic coupling layer. The TSC was carbon fiber reinforced epoxy with 180°C curing temperature and the coupling layer material was Polyvynilbutyral (PVB), which is a semi-crystalline thermoplastic polymer with melting temperature in the 150°C-170°C range. PVB is a low-cost polymer characterized by excellent binding and film forming ability and adhesion to many surfaces [23]. It has usually been used as an interlayer material in the manufacture of laminated glass for its high mechanical strength and optical clarity [24] but never as a coupling layer for welding of TSCs. A remarkable feature of the PVB coupling layer is that, owing to its low viscosity during the first stage of the co-curing process, i.e. before gelation of the epoxy resin, the PVB resin was able to partially penetrate in the first layer of the carbon-epoxy composite. This resulted in an alternative procedure to generate macromechanical interlocking between the coupling layer and the underlying composite. The CF/epoxy adherends with the PVB coupling layer were subsequently welded and their strength and failure mechanisms were analyzed. Two welding techniques novel for the intended application, namely induction and ultrasonic welding, and two different coupling-layer thicknesses were comparatively evaluated.

2. Experimental

A carbon fabric/epoxy prepreg supplied by Hexcel 3501-6 with a fiber volume content of 58% was used. 14 plies of CF/epoxy prepreg were stacked adding as a last ply a poly-vynil-butyral (PVB) film (Mowital, supplied by Kurakay) with two different thicknesses (75 and 250 µm). PVB is a random terpolymer containing butyral and hydroxyl side groups with a small amount of acetate units [25]. The hydrophobic vinyl butyral units provide elasticity and toughness while the hydrophilic vinyl alcohol units confer high adhesion to inorganic materials [26]. PVB has a glass transition temperature between 60 °C and 85 °C and a melting range between 150 °C and 170 °C.

The CF/epoxy/PVB stacks were cured in a hot platen press at 180 °C and 2 bar for 1 hour with no vacuum bag, resulting in laminates with a final thickness of 2.7 mm. Subsequently, the CF/epoxy/PVB cured adherends (cut from the CF/epoxy/PVB laminates) were welded in a single lap configuration as schematically shown in Figure 1. Two welding processes, namely induction and ultrasonic welding, were applied to join two adherends with a 250 µm-thick coupling layer (referred to as Ind250 and Us250 joints, respectively) and two adherends with a 75 µm-thick coupling layer (referred to as Ind75 and Us75 joints, respectively).

An induction welding setup (Fig. 2a), operating at 600 kHz and 220 V, tuning the power between 1 and 2 kW and using a coil speed of 2 mm/s, was used [27]. Heating was produced within the conductive patterns present in the carbon fiber fabric without adding any additional conductive mesh at the welding interface. The feedback control on the electric current flowing in the coil was based on a surface temperature measurement through an optical pyrometer. To avoid polymer degradation and composite delamination, the temperature of the upper surface of the joint was kept below a defined value, thanks to an air flow from a cooling nozzle. A cooled cylinder applied the consolidation pressure. Induction welded panels, with a total welded length of 220 mm and an overlap width of 20 mm, were produced in this set up. Out of each welded panel (one Ind250 and one Ind75 panel), 10coupons were cut to a 20 mm width using a diamond saw.

Ultrasonic welding of individual coupons was performed using a Rynco Dynamic 3000 ultrasonic welder and a welding jig custom-designed and built at TU Delft (Fig. 2b) described in [28]. A displacement-controlled welding process was used with the following welding parameters: 1500 N welding force, 86.2 µm peak-to-peak amplitude, 0.25 mm travel (i.e. displacement 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 an experimental procedure explained in[29]. The coupons, which were water jet cut from the cured CF/epoxy/PVB laminates to the dimensions 25 mm x 101 mm, were welded with an overlap length of 12.5 mm. Two different welding procedures were used for the different types of laminates considered in this study. In the case of the laminates with a 75 µm-thick PVB coupling film, i.e. Us75 joints, a 250 µm-thick flat energy director was placed between the samples to be welded prior to the welding process to ensure preferential heat generation at the welding interface [14]. In the case of the laminates with a 250 µm-thick PVB coupling film, i.e. Us250 joints, successful heat concentration at the welding interface could be achieved without using an energy director. Average vibration times for the welding of samples with a 75 and 250 µm-thick PVB welding surface were approximately300 ms and 250 ms, respectively.

Single lap shear tests were performed according to the ASTM D1002 standard to evaluate the strength of the induction and ultrasonic welded joints. A Zwick/Roell 250 kN testing machine and a MTS Insight 100 dynamometer were used for the ultrasonic and induction welded joints, respectively, at a crosshead speed of 1.3 mm/min. The apparent lap shear strength (LSS) of the joints was calculated as the maximum load divided by the total overlap area. It must be noted that the overlap area of the Ind250 and Ind75 joints was 20 x 20 mm², whereas the overlap area of the Us250 and Us75 joints was 25 x 12.5 mm²according to coil and sonotrode dimensions, respectively. A minimum of five samples were tested per type of weld configuration (i.e. thickness of the PVB coupling film) and type of welding process used in this study.

The thermal properties of the PVB film and of the CF/epoxy/PVB cured laminates were characterized by differential scanning calorimetry (DSC) using a 822e DSC (Mettler Toledo). Dynamic DSC scans from 25 °C to 250 °C at 10 °C/min on 10 mg of samples were performed in nitrogen atmosphere.

Cross section microscopic analysis of the as-manufactured laminates and of the welded samples was performed using a Zeiss optical microscope. It must be noted that the micrographic samples were grinded (up to 5000 grit number) but could not be subjected to the last stages of polishing because the PVB-rich weld line was dissolved by the ethanol present in the polishing solution. Post-mortem fractographic analysis of the welded samples was performed using a Jeol JSM-7500F scanning electron microscope (SEM).

3. Results

3.1 PVB-epoxy interaction

Fig. 3 shows a representative cross section optical micrograph of a sample from one of the asmanufactured CF/epoxy/PVB laminates. As observed in this micrograph, the CF/epoxy composite

featured significant porosity which is believed to result from the fact that the laminates were manufactured in a hot platen press instead of an autoclave, owing to restrictions in the equipment available for this research. The relatively low initial quality of the CF/epoxy/PVB laminates did however not have a major impact in the results of this research, as it will be shown later. The PVB coupling layer could also be seen in Fig. 3. Even though there is no observable color difference between the PVB resin and the microscopy embedding resin, the presence of the PVB coupling layer could be inferred by differences in the scratches. Thickness measurements taken on that micrograph from the outermost fibers until the external surface of the coupling layer lied between 200 and 280 µm, for an initial coupling layer thickness of 250 µm.

Fig. 4 presents a detail of the interface between CF/epoxy and PVB. Owing to the fact that the PVB and the epoxy resins displayed distinct shades of grey under the optical microscope, Fig. 4 clearly shows presence of PVB resin within the first CF/epoxy layer, even partially surrounding some of the carbon fiber bundles. This partial penetration of the PVB coupling layer in the CF/epoxy laminate can be regarded as a source of macro-mechanical interlocking between those two elements. Most likely, penetration of the PVB resin occurred in an early stage of the curing cycle, in particular before reaching the gel point of the epoxy resin, enabled by the liquid state of the epoxy resin and a temperature-induced drop in the viscosity, and hence flow, of the PVB resin. The flow of the PVB resin could explain the fact that the measurements taken in Fig. 3 for the final thickness of the coupling layer fluctuated around its initial value.

In order to assess whether any additional, micro-mechanical, connection between PVB and the epoxy resin of the composite used in this study could be expected, the solubility of the two polymers was assessed by determining the solubility parameter, δ , which according to the Hansen theory [30], can be expressed as:

$$\delta^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \tag{1}$$

where δ_d is the contribution of dispersion forces, δ_p is the contribution of permanent dipoles, and δ_h

is the contribution of hydrogen bonding. The contributions to the solubility parameters of epoxy and PVB was calculated by the group contribution method[31]. The interaction radius, R_a , which expresses the solubility parameter "distance" between two materials based on their respective partial solubility parameter components, was calculated from [30]:

$$R_{a}^{2} = 4(\delta_{d,f} - \delta_{d,r})^{2} + (\delta_{p,f} - \delta_{p,r})^{2} + (\delta_{h,f} - \delta_{h,r})^{2}$$
(2)

where subscripts f and r refer to the PVB film and the epoxy resin, respectively. The epoxy matrix of the prepreg used for composite preparation is supplied by Hexcel with the commercial name of 3501-6. The epoxy composition is not specified in the technical datasheet, where only the curing temperature of 180 °C is recommended. Therefore, the solubility parameters of the epoxy matrix cannot be exactly calculated, but only estimated. In the present case, for the calculation of solubility parameters, the most widely used epoxy oligomers for structural applications have been considered. They are diglycidyl ether of bisphenol A (DGEBA) and tetraglycidyl 4,4'-diaminodiphenylmethane (TGDDM). As an hardener, the aromatic amine diamino diphenyl methane (DDM) enabling the epoxy cure at 180 °C has been considered. The values of solubility parameters for DGEBA/DDM and TGDDM/DDM system and thermoplastic PVB polymer are reported in Table 1. As a comparison, the solubility parameters estimated by Hansen [30] for a hot curing epoxy have been also reported in Table 1. The obtained value of RA for the PVB-DGEBA/DDM, PVB-TGDDM/DDM and PVB-epoxy hot curing systems are 6.37, 8.19 and 7.5 ^{J1/2}/cm^{3/2}, respectively. Even if the solubility parameters are close, solubility depends not only from thermodynamics aspects as measured by the solubility parameters, but also by kinetic aspects related to the interfacial contact area and the molecular weight of solute and solvent.

Additionally, the DSC results confirmed the absence of solubility of PVB in the epoxy matrix of the composite laminates. As shown in Fig. 5, the glass transition temperature of PVB, calculated as the inflection point in the heat flux curve, is 71 °C. The dynamic DSC scan revealed that the CF/epoxy-PVB specimen (extracted from the upper part of the laminate with the PVB modified surface)

presents two values of T_g , around 69 °C and 192 °C, which indicates the presence of two distinct phases corresponding to the T_g of the two separate materials, i.e. PVB and epoxy matrix, respectively.

3.2 Morphology of welded TSC-TSC joints

Cross-section optical microscopy was used to study the morphology of the welded TSC-TSC joints. Special attention was paid to the thickness of the weld line in the different types of welded joints produced in this study. Fig. 6 displays representative cross section micrographs for the Ind250 and Us250. Fig.6 clearly shows that the induction welded joints featured a significantly thicker weld line than the ultrasonically welded joints. Measurements taken on the micrographs indicated that the weld-line thickness in the Ind250 joints ranged between 340 and 420 µm whereas the weld-line thickness in the Us250 joints ranged between 120 and 160 µm (see Table 2). The fact that in both types of welded joints the weld lines were thinner than the two initial adjacent PVB layers evidences the occurrence of flow of the PVB resin in both types of welding processes. However, much more flow occurred in the ultrasonic welding process. In the case of the induction welded samples some extra porosity, additional to the one present in the laminates before welding (Fig. 3), was observed, which could be caused by possible deconsolidation induced by through-the-thickness temperature gradients during the welding process. This extra porosity did not seem to be present in the ultrasonically welded samples most probably owing to effective prevention of thermal degradation through ultra-short heating times [11]. The presence or absence of this extra porosity is however hard to prove owing to the relatively low initial quality of the CF/epoxy/PVB laminates.

Cross section micrographs of the Ind75 and Us75 joints showed again a thicker weld line for the induction welded joints (see Table 2 and Fig.7). In the case of the ultrasonic welded joints, despite the fact that an extra layer of 250 μ m-thick PVB resin, i.e. energy director, was added in between the adherends prior to the welding process, the thickness of the weld line was found to be similar to that of the resin-rich areas in the composite layers. In the case of the induction welded joints, the

measured weld line thickness indicated that flow of the PVB resin during the welding process was very limited.

3.3 Mechanical properties of the TSC-TSC welded joints

The results of the lap shear tests are shown in Table 3. The LSS values yielded by the Ind250, Us250 and Us75 samples were somewhat similar to each other (Ind250: 22.4 ± 1.4 MPa, Us 250: 27.9 ± 1.2 MPa, Us75: 24.3 ± 2.9 MPa). The LSS values yielded by the Ind75 welded samples were however around 40% lower than the LSS of the Ind250 samples (Ind75: 13.0 ± 1.0 MPa). It must be noted than a more in-depth comparison among the LSS values presented in Table 3 is not possible primarily owing to the differences in weld line thicknesses shown in the previous section as well as the differences in overlap length between the Ind and Us welded samples. Such differences can be expected to cause changes in the stresses developed at the weld line during the single lap shear tests [33], [34] and to hence hinder one-on-one comparisons, for which detailed calculations of the different stress fields, out of the scope of this paper, would be necessary. However, the LSS values presented in Table 3 are useful as some preliminary information of the quality of the CF/epoxy welds. This information can be widened though a detailed study of the failure modes in the different types of samples.

3.4 Failure of welded TSC-TSC joints

Cross-section microscopic analysis of the Ind250 joints showed that in that type of joints failure occurred around two layers deep within one of the adherends, leaving behind a fairly intact weld line as seen in the micrograph in Fig.8. The corresponding fracture surfaces were hence relatively flat and showed typical features of brittle resin (i.e. epoxy resin) failure, as shown in Fig.9. They also evidenced significant porosity. This type of failure, i.e. delamination of the adherend, could be caused by the combined effect of relatively high peel stresses during the single lap shear test owing

to secondary bonding (which increases with increasing weld line thickness) and the relative low quality, and hence low peel strength, of the laminate.

In the ultrasonic welded US 250 joints failure was predominantly found to occur within the weldline and/or the first composite layer in one of the adherends. The cross-section micrographs in Fig. 10 show an example of failure in the first composite layer. Fig. 11 shows however a representative fracture surface of a sample that failed at the weld line. It shows uniformly distributed resin patches with significant plastic deformation (presumably PVB resin) and patches of seemingly bare fibers. Detailed observation of the fracture surface in the SEM shows peeling of resin which leaves behind bare carbon fibers (fiber-resin debonding). The high amount of plastic deformation in this resin as well as the fact that no resin residues stay on the fibers, indicate that this is the PVB resin that during the curing cycle wetted the carbon fiber bundles adjacent to the weld line.

In the case of the Ind75 joints, failure was found to occur at the welding overlap. The fracture surfaces (an example of which is shown in Fig. 12) indicate that approximately half of the overlap experienced debonding between the two adjacent PVB layers, most probably caused by an insufficient welding temperature or pressure in that area. The other half showed resin-rich patches and patches of seemingly bare carbon fibers. SEM inspection of that area revealed the occurrence of fiber-resin debonding, presumably between PVB resin and carbon fibers, plastic deformation of the PVB resin and local debonding between adjacent PVB layers due to insufficient welding. The fact that approximately only half of the overlap experienced welding to a certain extent explains the significantly lower LSS values yielded by the Ind75 samples. Nevertheless, if only the actual welded area were considered to calculate the LSS of these samples, the final value would be close to the values yielded by the other types of samples.

The Us75 joints also showed failure at the weld line with, however, a fully welded overlap. The corresponding fracture surfaces (an example of which is shown in Fig. 13) contained PVB resinrich and bare fiber patches. The PVB resinrich patches, which were visibly thinner than the ones in

Figs. 8 (Us250 joints) and 10 (Ind75 joints), showed extensive plastic deformation most likely owing to their small thickness. Fiber-resin debonding was as well observed in this case. Similarly to the other cases shown before, bare carbon fibers could be found on the fracture surfaces as displayed in Figs. 13 and 14. However, the fracture surfaces of the Us75 joints also featured some areas where the carbon fibers had a rough texture on their surface as shown in Fig. 14.

4. Discussion

The cross-section micrographic analysis of the as-manufactured CF/epoxy/PVB laminates showed that the PVB resin locally displaced the epoxy resin during co-curing partially penetrating in the first composite ply and partially wetting the fiber bundles. This could as well be inferred from the fracture surfaces of the welded joints which failed at the weld line/first ply. They featured fiber-matrix debonding mostly resulting in bare carbon fibers, which is a typical failure mode in composites involving a thermoplastic matrix [35] owing to the typically low adhesion between thermoplastic polymers and reinforcing fibers [36].

The lap shear strength (LSS) of the welded joints that failed at the weld line/first ply was around 25 MPa. A very wide range of LSS is available in the literature, depending on the type of epoxy resin used, the curing cycles, the eventual presence of a reinforcement into the adhesive: the reported values of LSS with CF/epoxy range between 11 and 30 MPa [37],[5], [38], [39]. The main failure mode was fiber-matrix debonding between the PVB and the reinforcing carbon fibers within the first composite ply accompanied by significant plastic deformation of the PVB resin. This type of failure points towards mechanical interlocking between PVB and the reinforcing fibers as the main basis for the strength of the welded joints. The relatively good adhesion between the epoxy and PVB, as indicated by their partial solubility values, could as well play a role in the strength of the welded joints. The PVB weld line showed however very low resistance to ethanol, which indicates that the chemical resistance could be a limiting factor for this type of joints. Owing to the

relatively low glass transition and melting temperature of PVB, the maximum operational temperature for this type of joints would be around 80°C.

Both the type of welding processes and the thickness of the coupling layer were found to have an effect on the final thickness of the weld line. Obviously, thicker coupling layers resulted in thicker weld lines, which were in any case thinner than the initial PVB-rich area owing to resin squeeze-out during the welding processes. The resin squeeze-out was much more significant in ultrasonic welding for which only around 25% of the original PVB material initially at the welding interface (coupling layers and energy director, when applicable) remained in the weld line. The different weld line thicknesses were found to have an effect on the type of failure of the welded joints. In particular the welds with the thickest weld line (around 0.4 mm-thick weld line in the Ind250 joints)experienced premature interlaminar failure in the CF/epoxy composite away from the weld line. This could most likely be due to higher peel stresses resulting from higher secondary bending [33] combined with the reduced quality of the composite laminate. The use of the thicker coupling layers was however found to be beneficial for the welding processes. In the case of ultrasonic welding, the thicker coupling layer enabled successful welding without the need of an extra energy director. In the case of induction welding, the overlap area was completely bonded when the thicker thermoplastic film was used. This can be considered the main reason of higher LSS for these samples.

The reasons why thin coupling layers hindered the creation of complete induction welded joints are unknown to the authors and need further research. Finally, the fracture surfaces of ultrasonically welded samples with thin coupling layers showed some unusual features that might be indicating some sort of thermal degradation of the fiber sizing during the welding process. This is however a preliminary hypothesis and needs to be investigated further.

Conclusions

The research presented in this paper focused on the welding of CF/epoxy composites through a PVB thermoplastic coupling layer. Two different coupling layer thicknesses were investigated as well as two different welding processes, induction and ultrasonic welding. The following main conclusions were drawn from the results of this research:

- Macro-mechanical interlocking between the PVB coupling layer and the CF/epoxy composite was achieved through partial penetration of the PVB resin in the first composite layer during co-curing. This is believed to have been facilitated by the coexistence of low-viscosity PVB and liquid epoxy in the early stage of the curing cycle, i.e. before reaching the gel point of the epoxy matrix.
- The resulting CF/epoxy to CF/epoxy welded joints (through the PVB coupling layers) yielded around 25 MPa apparent lap shear strength, which successfully lies within the typical range of strength values for CF/epoxy adhesively bonded joints. The main failure mechanism was fiber-matrix (PVB) debonding, which points at mechanical interlocking between PVB and the carbon fibers as the main responsible for the strength of the joints.
- Both induction and ultrasonic welding processes resulted in adequately strong welded joints. The main difference between the two processes was a much higher amount of resin squeezed out of the interface during ultrasonic welding, which resulted in thinner weld lines. Thicker weld lines were found to induce premature failure within the CF/epoxy laminate, away from the weld line. In those cases, the actual strength of the welded joint could not be tested.
- Thicker PVB coupling layers were found to ease the welding process. In the case of ultrasonic welding no extra energy director was needed and in the case of induction welding a fully welded overlap was successfully achieved.

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Figure Captions

Figure 1: Schematic representation of the TSC-TSC joints before welding.

Figure 2: a) Induction welding setup and b) ultrasonic welding set up: (1) sonotrode, (2) vertically sliding platform for clamping of top adherend, (3) clamping of top adherend, (4) clamping of bottom adherend.

Figure 3: Cross section optical micrograph of sample from CF/epoyx/PVB laminate with 250 µmthick PVB coupling layer and detail showing position of outer surface of coupling layer (dotted line). Position of outer surface of coupling layer is also indicated in main figure with an arrow. Small rectangle indicates approximate location of detail in Fig. 4.

Figure 3: Left: Detail in cross section of adherend with a 75µm-thick PVB layer that shows wetting of the outermost fibers in a bundle and penetration of PVB resin between the warp and weft bundles within first ply of CF/epoxy composite. Right: The boundaries between PVB and epoxy resin have been highlighted with dotted line.

Figure 5:Dynamic DSC scan of PVB and CF/epoxy-PVB at 10 °C/min in nitrogen atmosphere.

Figure 6: a) Cross section micrograph in the centre of Ind250 joint; b)cross section micrograph in the centre of Us250 joint.

Figure 7: a) Cross section micrograph in the centre of Ind75 joint; b) Cross section micrograph in the centre of Us75 joint. The arrow indicates the position of the weld line.

Figure 8: Cross section micrograph of tested coupon (Ind250) showing delamination failure in one of the adherends. The arrow indicates the position of the weld line.

Figure 9: a) Representative fracture surface (Ind250); b) SEM detail showing features of brittle resin failure as well as void imprints.

Figure 10: a) Cross section micrograph of welded adherend after testing (Us250) showing failure in the composite ply adjacent to the weld line; b) Detail at higher magnification showing damaged fiber bundle adjacent to the weld line. (Arrows indicate position of weld line).

Figure 11: a) Representative fracture surface of Us250 coupon failed at the weld line showing resin-rich patches with significant plastic deformation and patches with seemingly bare fibers; b) SEM detail showing fiber-resin debonding.

Figure 12: a) Representative fracture surface of induction welded joint (Ind75); b) SEM detail showing debonding between fiber-resin debonding.

Figure 13: a) Representative fracture surface of ultrasonic welded joint (Us75); b) SEM detail showing fiber-resin debonding and extensive plastic deformation of the resin.

Figure 14: SEM detail of same Us75 fracture surface showing: a)bare carbon fibers and b) carbon fibers with rough texture on their surface.











































b)

2<u>0 µm</u>

















Polymer	δ _D (MPa) ^{0,5}	δ _P (MPa) ^{0,5}	δ _H (MPa) ^{0,5}	δ (MPa) ^{0,5}	
DGEBA/DDM	18.57	3.03	7.83	20.38	
TGDDM/DDM	20.72	2.17	8.20	22.38	
Epoxy hot curing	18.30 [30]	12.30 [30]	9.70 [30]	24.09 [30]	
PVB	18.74	9.33	8.72	22.68	

Table 1: Solubility parameters for the investigated polymer systems.

Welding process	Initial thickness PVB layer (µm)	Weld line thickness range	
		(µm)	
Induction	250	320 - 420	
Ultrasonic	250	120 - 160	
Induction	75	130 – 180	
Ultrasonic	75	100*	

Table 2: Measured weld line thicknesses for induction and ultrasonic welded joints.

* with the addition of a 250 µm-thick flat PVB energy director between the samples to be welded.

250 μm film 75 μm film Induction welded(20x20 mm ² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)		Induction welded(20x20 mm ² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap) 2000 mm ² 2000 mm ²	Induction welded(20x20 mm ² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3± 2.9 (25 x 12.5 mm ² overlap)	Induction welded(20x20 mm ² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap) 27.9 ±1.2 24.3 ± 2.9	Induction welded(20x20 mm² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3± 2.9 (25 x 12.5 mm² overlap) (25 x 12.5 mm² overlap)	Specimen Type	LSS ± Stdev (MPa)	
Induction 22.44 ± 1.4 13.0 ± 1.0 welded($20x20 \text{ mm}^2$ overlap) 27.9 ± 1.2 24.3 ± 2.9 (25 x 12.5 mm^2 overlap) $(25 \times 12.5 \text{ mm}^2 \text{ overlap})$	Induction welded($20x20 \text{ mm}^2$ overlap) 22.44 ± 1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ± 1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap) \mathbf{V}	Induction welded($20x20 \text{ mm}^2$ overlap) 22.44 ± 1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ± 1.2 24.3 ± 2.9 (25 x 12.5 mm^2 overlap) $2000000000000000000000000000000000000$	Induction welded(20x20 mm² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3± 2.9 (25 x 12.5 mm² overlap) 2000 mm² 2000 mm²	Induction welded(20x20 mm² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm² overlap) (25 x 12.5 mm² overlap)	Induction welded(20x20 mm² overlap) 22.44 ±1.4 13.0 ± 1.0 Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm² overlap) (25 x 12.5 mm² overlap)		250 μm film	75 μm film
Ultrasonic welded 27.9 ±1.2 24.3± 2.9 (25 x 12.5 mm ² overlap)	Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)	Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)	Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)	Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)	Ultrasonic welded 27.9 ±1.2 24.3 ± 2.9 (25 x 12.5 mm ² overlap)	Induction welded(20x20 mm ² overlap)	22.44 ±1.4	13.0 ± 1.0
(25 x 12.5 mm ² overlap)	(25 x 12.5 mm ² overlap)	(25 x 12.5 mm ² overlap)	(25 x 12.5 mm ² overlap)	(25 x 12.5 mm ² overlap)	(25 x 12.5 mm ² overlap)	Ultrasonic welded	27.9 ± 1.2	$24.3{\pm}~2.9$
						$(25 \text{ x } 12.5 \text{ mm}^2 \text{ overlap})$, C
							2	
							AP	
				\mathbf{G}^{-}				
				C .				

 Table 3. Results of single lap shear tests.