HYBRID WELDING OF CARBON-FIBRE REINFORCED EPOXY BASED COMPOSITES

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Keywords: thermoset composites, thermoplastic composites, hybrid welding, ultrasonic welding, induction welding

Abstract
Two fusion bonding techniques typically used for thermoplastic matrix composites, such as ultrasonic and induction welding, were applied for joining thermosetting matrix composites. A proper modification of the composite layup was proposed, i.e. a thermoplastic film of Poly-vinyl butyral (PVB) was added as a last ply in the stacking sequence of carbon fibre epoxy prepregs, typically used in aeronautical applications. After matrix curing, an intermingled thermoplastic-thermoset polymer zone at the surface of composite was obtained. The composite containing the thermoplastic film was used for fabrication of single lap joints by ultrasonic and induction welding, exploiting the melting of the thermoplastic film. The joint properties and the chemical compatibility and adhesion of PVB to the carbon fiber-epoxy prepregs were studied by mechanical testing and microstructural analysis.

1. Introduction

Thermoplastic composites (TCPs) offer several advantages compared to thermoset composites (TSCs). Besides the reduction of processing times and the enhanced possibilities for recycling, a significant benefit is presented by the possibility of automated joining of thermoplastic composite parts . Since assembly of fiber reinforced composites roughly represents 50% of the manufacturing costs of an aeronautic structure, faster and cheaper assembly technologies strongly attracts the interest of industry [1]. Fusion bonding techniques eliminate the well known drawbacks of the traditional joining techniques for composite materials, like stress concentration and intense labor associated to mechanical joining or extensive surface preparation and long curing cycles related to adhesive bonding [2].
Welded joints present several advantages with respect to bolted joints, such as weight reduction, increased mechanical performance due to the more homogeneous load transmission and short processing cycles [3-4]. Among fusion bonding techniques for TPCs, induction welding and ultrasonic welding are the most promising since they can provide adequate mechanical properties and several processing advantages, such as high speed, efficiency and repeatability [5-7].

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

Ultrasonic welding involves the use of high frequency mechanical waves to soften or melt the thermoplastic matrix at the joint line. The parts to be joined are held together under pressure and subjected to ultrasonic vibrations applied by a welding horn, usually at a frequency of 20 or 40 kHz. In comparison to other joining techniques, ultrasonic welding is characterized by a low energy input, consequently low temperatures in the welding zone, very short welding times and ease of automation [5]. Induction welding is based on electromagnetic induction heating for a contact free heating and a subsequent adhesion by pressure. When an electrically conductive workpiece is placed close to a coil crossed by an alternating current, which generates an alternating time-variable magnetic field, eddy currents are induced in the workpiece. A conductive metal mesh (a susceptor) must be used at the welding interface when using glass fibers while carbon fiber composites can be heated without using any additional susceptor, thanks to the conductive properties of carbon fibres [8-9].

Till now, welding techniques have been limited to joining continuous fiber-reinforced thermoplastic composites since thermoset matrices cannot melt due to their crosslinked molecular structure. The possibility of applying fusion bonding technologies to the joining of TSC structures is attracting growing attention for the potential reduction in assembly times, assembly costs and weight as compared to mechanical fastening [9]. Even if some developments in hybrid welding were reported since the end of nineties [10-11] a renewed interest for joining thermoset matrix composites using a thermoplastic layer at the surface of the thermoset composite recently appeared [1]. A thermoplastic polymer film introduced as a last ply before the cure of the thermosetting composite laminate provides a meltable layer at the surface of the laminate [12].

In this work, the applicability of ultrasonic and induction welding for joining carbon reinforced epoxy composites has been investigated. The thermoset composite layup is modified by the addition of a thermoplastic film as a last ply in the stacking sequence. Due to co-curing, an intermingled thermoplastic-thermoset polymer zone at the surface of composite is obtained, which can be used for further assembly operations by welding. During ultrasonic or induction welding, the melting of the thermoplastic film has been exploited to join the composite parts.

2. Experimental

The materials used in this study was a carbon fabric/epoxy prepreg supplied by Hexcel with the commercial name 3501-6 and a fiber volume content of 58%. 14 plies of CF/epoxy prepreg were stacked adding as last ply a thermoplastic film, as schematically shown in Fig. 1. The used thermoplastic film was poly-vynil-butyral (PVB) with the commercial name Mowital, supplied by Kurakay with two different thicknesses (45 and 250 μm). The laminate was cured in a press at 180 °C and 2 bar for 1 hour allowing the development of a bond between the composite plies and the thermoplastic film. The cured laminate had a final thickness of 2.7 mm.

An induction welding set-up (Fig. 2a), designed and developed by CETMA and SINERGO (Italy), was used [8]. Induction welding was performed at 600 kHz and 220 V tuning the power between 1 and 2 kW and using a coil speed of 2 mm/s. Heating was produced within the conductive patterns present in the carbon fibre 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.

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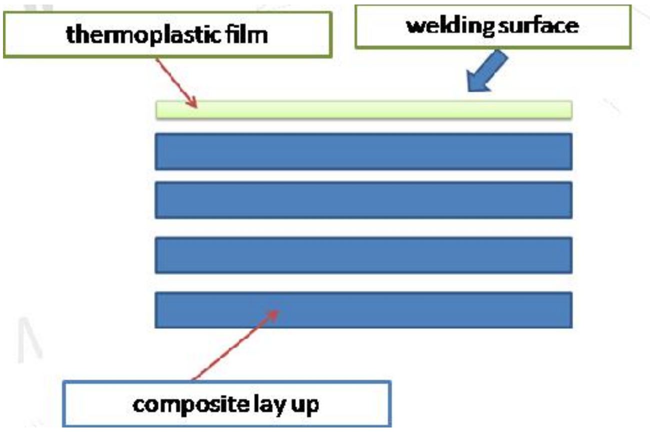

Figure 1. Schematic representation of the stacking sequence before curing.

Individual test samples were ultrasonically welded using a Rynco Dynamic 3000 ultrasonic welder and a welding jig custom-designed and built at TU Delft (Fig. 2b). 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 the procedure explained in [13]. Two different welding procedures were used for the two different types of laminates considered in this study. In the case of the laminates with a 45 μm-thick PVB welding surface, a 0.25 mm-thick flat energy director was placed between the samples to be welded to ensure preferential heat generation at the welding interface [14]. In the case of the laminates with a 250 μm-thick PVB welding surface successful heat concentration at the welding interface could be achieved without an energy director. Average vibration times for the welding of samples with a 45 μm-thick PVB welding surface were around 300 ms, and 250 ms for the samples with a 250 μm-thick PVB welding surface.

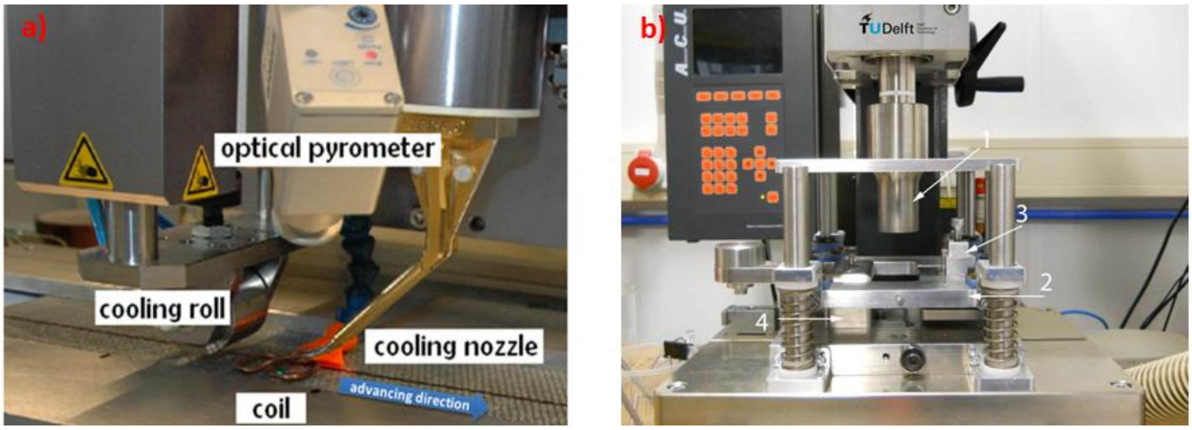

Figure 2. a) Induction welding setup and b) ultrasonic welding set up: (1) sonotrode, (2) vertically sliding platform for clamping of top adherend, (3) clamping of top adherend, (4) clamping of bottom adherend.

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Single lap shear tests were performed according to the ASTM D1002 standard to evaluate the strength of the composite joints welded by means of induction or ultrasonic welding. A Zwick/Roell 250 KN testing machine and a MTS Insight 100 dynamometer were used for the ultrasonic and induction welded joints, respectively. In both cases the overlap area (25 x 12.5 mm^2) and the crosshead speed (1.3 mm/min) were the same. The apparent lap shear strength (LSS) of the joints was calculated as the maximum load divided by the total overlap area.

The thermal properties of the thermoplastic film and the cured laminate was characterized by differential scanning calorimetry using a 822e DSC (Mettler Toledo).

The microstructure of the samples was observed by a scanning electron microscopy (SEM) using a Zeiss EVO 40 SEM instrument at variable pressure operating with a voltage of 20KV and a JSM-7500F Scanning Electron Microscope.

3. Results

In order to achieve a strong joint, different challenges need to be overcome both during curing and welding. First of all, a good adhesion between the thermoset and the thermoplastic polymer was obtained before welding through a proper choice of the thermoplastic film, accounting for its properties as described by the solubility parameter and thermal transitions. The solubility parameters of different thermoplastic films and composite matrix were compared in a previous work [15] as well the cure temperature of the epoxy matrix and the glass transition temperature (T_g) of the films. The DSC characterization confirmed the amorphous nature of PVB with a T_g of 70 °C.

During welding, the bond strength of welded components mainly depended on: i) the welding temperature dictated by the thermoplastic film transitions; ii) the glass transition of the thermosetting composite matrix and its degradation temperature; iii) the surface damage, thermal stresses and crack generation.

The solubility of poly-vynil-butyral in the epoxy matrix of the composite joint was analyzed. According to the Hansen theory [16], the solubility parameter, δ, was determined as the sum of three contributions:

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

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 thermoplastic film was evaluated by the group contribution method [17]. 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 [16]:

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

where subscripts f and r refers to the PVB film and the epoxy resin, respectively. The obtained value of Ra for the PVB-epoxy system was 6.37 $J^{1/2}/cm^{3/2}$, indicative of a partial solubility of PVB in epoxy resin. A complete solubility is not desired since it can lead to dissolution of the film in the resine during curing before gelation. However a good adhesion between the thermosetting and thermoplastic resins is promoted by similar solubility parameters. The properties of PVB were considered a compromise between these opposite requirements.

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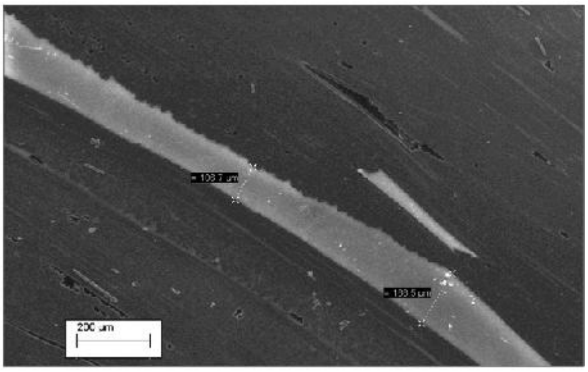

Figure 3. SEM micrographs of a carbon/epoxy joint welded by means of a PVB film.

The microstructural analysis carried out by scanning electron microscopy (SEM) on induction welded specimens is reported in Fig. 3. The thickness of PVB, initially of 250 μm, was reduced to about 140 μm as a consequence of solubilization in the resin occurred during curing. Moreover, mechanical interlocking was observed in Fig. 3 at the interface between the PVB film and the TS composite, thus confirming partial solubilization of PVB in the liquid matrix and related wetting of reinforcement fibers, in any case leading to an improvement of the adhesion between the thermoplastic and thermosetting interface.

The results of lap shear test, reported in Table 1, evidenced a good adhesion strength. The LSS was high, very close to other literature results on CF/epoxy [18]. In particular, the ultrasonically welded samples presented a higher LSS compared to induction welded samples. This difference was very significant when the thinner PVB film was used. This deserves further studies considering that an additional film of 0.25 mm was added during ultrasonic welding and that the mentioned partial solubility of PVB in the epoxy matrix could lead to a very small thickness of PVB at the welding interface after laminate curing. A higher LSS observed during ultrasonic welding can be also related to lower maximum temperature reached during welding which prevent epoxy resin degradation. This is a consequence of: (a) the fact that viscoelastic heating is mostly generated around the T_g of the PVB thermoplastic resin, which is relatively low, and (b) the very low heating times (under 300 ms) prevented matrix degradation [9].

Table 1. Results of single lap shear tests.

Specimen Type	LSS (MPa)	
	250 μm film	45 μm film
Induction welded	21.74 ±1.3	5.4 ± 1.3
Ultrasonic welded	27.9 ±1.2	24.3 ± 2.9

Induction and ultrasonically welded specimens with a 250 μm-thick PVB welding surface showed two different types of failure: cohesive failure of thermoplastic film and interlaminar in the CF/epoxy composite (see Fig. 4). In the case of ultrasonically welded specimens with a 45 μm-thick PVB welding surface, the only type of failure observed was cohesive failure, as shown in Fig. 5. The differences in failure type and lap shear strength between induction/ultrasonically welded samples with 250 μm-thick PVB welding surfaces and ultrasonically welded samples with 45 μm-thick PVB welding surfaces could potentially be attributed to a smoothening of the stress peaks within the

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relatively thicker PVB interface [19] in the former welded joints allowing for the critical interlaminar stress in the adherends to be reached before the critical stress in the PVB weldline.

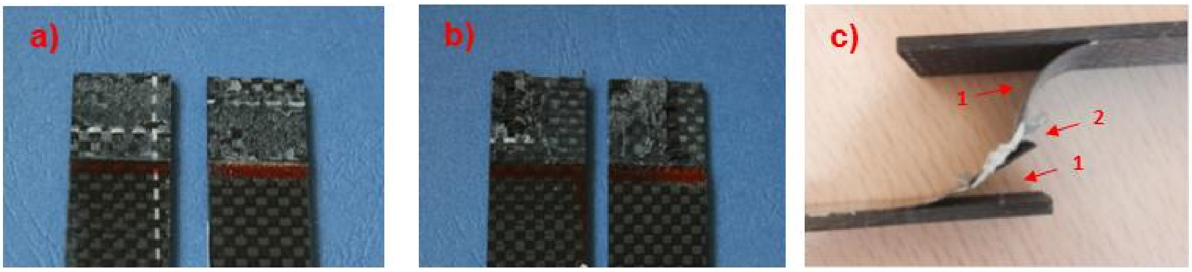

Figure 4. Representative failure types in welded CF/epoxy samples with 250 μm-thick PVB: a) cohesive failure in induction welded sample, b) interlaminar failure in the composite in the induction welded sample, c) ultrasonically welded sample showing interlaminar failure (1) and cohesive failure (2) simultaneously.

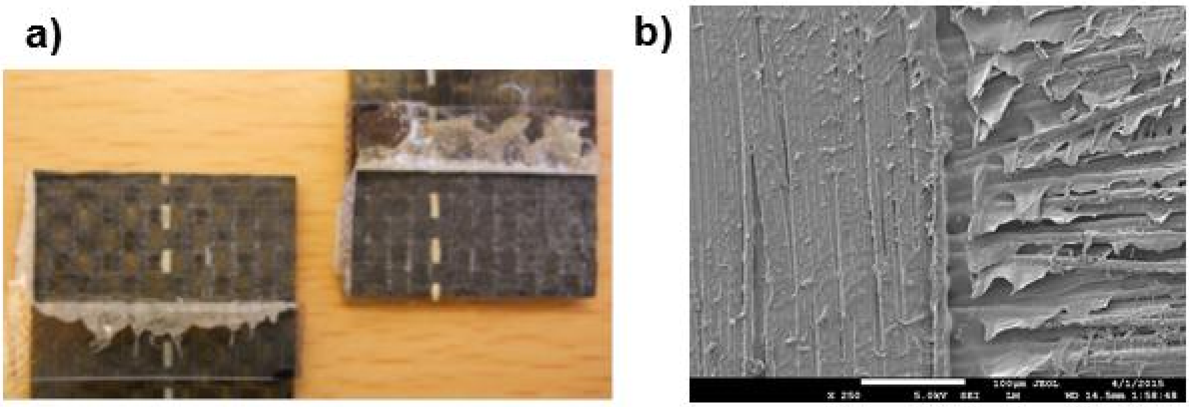

Figure 5. a) Representative fracture surfaces in ultrasonically welded CF/epoxy samples with 40 μm-thick welding surfaces and b) SEM details showing a resin-rich surface with signs of plastic deformation which indicates the occurrence of cohesive failure in this type of joints.

3. Conclusions

A new approach for the assembly of thermosetting composites by induction welding and ultrasonic welding was developed. Two thermosetting composites joints were obtained using carbon reinforced epoxy matrix composites characterized by a stacking sequence where, as a last ply, a PVB thermoplastic film with Tg 70 °C was added. The welded joint were characterized by single lap shear tests and microstructural analysis.
The complex relations among the material variables governing the final properties of the hybrid joint were highlighted. The compatibility and adhesion of an amorphous thermoplastic film to the epoxy matrix was related to the solubility parameter and the material transition temperatures. This study provided promising results, even though still preliminary and limited to the use of PVB at the interface. Additional studies are needed to correlate the properties of welded joints with the complex relationships among thermal properties, curing conditions and solubility parametres of the used materials.

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Acknowledgments

The authors would like to acknowledge Pierfrancesco Michele of University of Salento for the thermal and morphological characterization of the specimens.

References

[1] S. Deng, L. Djukic, R. Paton, and L. Ye. Thermoplastic–epoxy interactions and their potential applications in joining composite structures – A review. *Composites Part A: Applied Science and Manufacturing*, 68: 121-132, 2015.

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

[3] G. Scarselli G, E. Castorini, F.W. Panella, R. Nobile, and A. Maffezzoli. Structural behaviour modelling of bolted joints in composite laminates subjected to cyclic loading. *Aerospace Science and Technology*, 43: 89-95, 2015.

[4] F. Lionetto, R. Dell'Anna, F. Montagna, and A. Maffezzoli. Modeling of continuous ultrasonic impregnation and consolidation of thermoplastic matrix composites. *Composites Part A: Applied Science and Manufacturing*, 82: 119–129, 2016.

[5] I. Fernandez Villegas, L. Moser, A. Yousefpour, P. Mitschang, and H.E.N. Bersee. Process and performance evaluation of ultrasonic, induction and resistance welding of advanced thermoplastic composites. *Journal of Thermoplastic Composite Materials*, 26: 1007-1024, 2013.

[6] F. Lionetto, R. Dell'Anna, F. Montagna, and A. Maffezzoli. Ultrasonic assisted consolidation of commingled thermoplastic/glass fiber rovings. *Frontiers in Materials*, 2: 1-9, 2015.

[7] T. Bayerl, M. Duhovic,P. Mitschang, and D. Bhattacharyya. The heating of polymer composites by electromagnetic induction–A review. *Composites Part A: Applied Science and Manufacturing*, 57: 27-40, 2014.

[8] S. Pappadà, A. Salomi, J. Montanaro, A. Passaro, A. Caruso, and A. Maffezzoli. Fabrication of a thermoplastic matrix composite stiffened panel by induction welding. *Aerospace Science and Technology*, 43: 314-320, 2015.

[9] I. Fernandez Villegas and P.V. Rubio. On avoiding thermal degradation during welding of high-performance thermoplastic composites to thermoset composites. *Composites Part A: Applied Science and Manufacturing*, 77: 172-180, 2015.

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

[11] M. Hou, A. Beehag, and Q. Yuan. Welding techniques for polymer or polymer composite components. *International patent No. 011573*, 2004.

[12] I. Fernandez 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 ICCM-20, Copenhagen, Denmark*, July 19-24 2015.

[13] I. Fernandez 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.

[14] G. Palardy and I. Fernandez Villegas. Ultrasonic welding of thermoplastic composites with flat energy directors: influence of the thickness of the energy director on the welding process. *Proceedings of 20th International Conference on Composite Materials ICCM-20, Copenhagen, Denmark*, July 19-24 2015.

[15] S. Pappadà, A. Salomi, J. Montanaro, A. Passaro, F. Lionetto, and A. Maffezzoli. Induction welding for assembly of thermosetting matrix composites. *Proceedings of SAMPE Conference, Amiens, France*, September 15-17 2015.

[16] C.M. Hansen. *Hansen solubility parameters: a user's handbook*. CRC Press, 2007.

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

[17] A. Greco, F. Lionetto, and A. Maffezzoli. Processing and characterization of amorphous polyethylene terephthalate fibers for the alignment of carbon nanofillers in thermosetting resins. *Polymer Composites*, 36: 1096-1103, 2015.
[18] T. Beiss, M. Menacher, R. Feulner, G. Huelder, and T.A. Osswald. Vibration Joining of Fiber-Reinforced Thermosets. *Polymer Composites*, 31: 1205-1212, 2010.
[19] D.M. Gleich, M.J.L. van Tooren, and A. Beukers. Analysis and evaluation of bondline thickness effects on failure load in adhesively bonded structures. *Journal of Adhesion Science and Technology*, 15: 1091-1101, 2001

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