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The influence of epoxy adhesive toughness on the strength of hybrid laminate adhesive joints

R. J. C. Carbas^{1,2*} , E. A. S. Marques¹ and L. F. M. da Silva²

*Correspondence:

rcarbas@fe.up.pt

¹ Institute of Science and Innovation in Mechanical and Industrial Engineering (INEGI), Faculty of Engineering, University of Porto, Porto, Portugal
Full list of author information is available at the end of the article

Abstract

The use of composite materials in structural applications has significantly expanded in recent years. The transport industry accounts for an increasingly larger share of the final structural weight of vehicles, as manufacturers pursue improvements in fuel economy, lighter more efficient designs, and reduction of emissions. However, the delamination of adhesively bonded composite joints causes premature failure of the bond, inhibiting the use of its full potential and leading to inefficient and over-designed components. A hybrid composite metallic material technology is studied in this work, a method inspired in the fibre metal laminate concept, and which combines the best properties of FRPs and metal alloys. The hybrid composite-metallic adherends aims to increase the joint strength in the through thickness direction, minimise peel stresses and limit delamination. The objective of this work was to evaluate the performance of hybrid joints, bonded with different adhesives by comparing them against a reference joint using a conventional Carbon fibre reinforced polymer (CFRP) adherend. The joints were experimentally tested using a universal testing machine for a crosshead speed of 1 mm/min. Numerical models were developed, using the ABAQUS software, to study the behaviour of all joints studied. The numerical predictions of failure loads and modes were compared to the experimentally obtained results.

Keywords: Carbon fibre, Laminates, Mechanical properties, Finite element analysis, Mechanical testing, Anodizing

Introduction

The importance of composite materials in modern engineering cannot be overstated. Nowadays, composites represent a very important subset of materials for any designer/engineer, mainly due to their high specific stiffness or excellent strength to weight ratio, fatigue behaviour and corrosion resistance. This makes them very attractive for applications where weight is a major design constraint. However, joining composites is often a challenging proposition. In most cases, the joining will be guaranteed by adhesive bonding only, with adhesives taking on a critical structural role. The anisotropic nature of composites adds an extra layer of complexity to the use of these materials with possible out-of-plane/interlaminar loadings leading to delamination—a transversal failure mode due to peel stresses and poor bonding between the fibres and the polymeric matrix.

Delamination is seen as a major issue to tackle before a more representative use of composite materials in structural applications can occur, allowing to fully explore their inherent advantages [1, 2]. Currently, there are safety concerns about the detection of delamination cracks and, since studies show that the crack growth in CFRP is relatively high for low stress fatigue cycles, this could lead to unpredictable catastrophic failures of composite structures [3]. The initiation of the cracks themselves could be traced back to geometrical discontinuities, such as the holes for mechanical fasteners or even to impact damage [4]. In fact, the higher notch sensitivity of composites and their low shear strength is one of the reasons that adhesive joints became prevalent over mechanical connections for joining composites. The smooth uniform stress distribution observed along the bond translates into excellent joint performance and high fatigue resistance.

There are several methods available to mitigate the delamination issue [5], such as Z-pins or Z-anchoring (the introduction of thin pins, in the through thickness direction of the composite, holding the laminate plies together by a combination of adhesion and friction [6, 7]), 3D weaving (the reinforcement of interlaminar properties by the creation of complex three dimensional dry fibre preforms before applying the resin [8, 9]), stitching or tufting (embedding stitch threads through thickness direction, creating a bridging effect that will attenuate the delamination cracks [10, 11]), mixed adhesive joints or functionally graded adhesive joints (to decrease of stress concentration at the ends of overlap and increase of joint strength using a more flexible adhesive at the ends of the overlap to reduce the peel stresses in that critical section [12, 13]), and hybrid laminates (metal or polymeric laminates are used to reinforce the composite transversal properties [14–17]). The 3D weaving, stitching, braiding, tufting, z-pinning and z-anchoring techniques all successfully reduce delamination failure, but are quite laborious, which increases the cost of the final product [18].

The use of hybrid laminate concept is a technique which is simpler to implement industrially and can also be successful in delamination prevention. The authors showed previously that the delamination of composite joints can be avoided by reinforcing the composite adherends with thin metal [14–16] or polymeric layers [17]. The hybrid laminate technique allows to combine the mechanical properties of different laminates reinforced with composite.

The aim of this study was the evaluation of the joint performance of hybrid metal laminates bonded with different epoxy adhesives. The resultant hybrid joints were compared with joints using only CFRP adherends. The joints were tested using a universal testing machine at a rate speed of 1 mm/min. Numerical models were developed, using the ABAQUS software, to better understand the behaviour of all configurations under study. The numerical predictions of the failure loads and failure modes were compared to the experimentally obtained results.

Experimental details

Adhesive

Three different epoxy-based adhesives were considered for this work. These are the Araldite® AV138M with HV 998 hardener, 3M Scotch Weld AF 163 2K and Nagase Denatite XNR6852E-3. Although they are all epoxy-based structural adhesives they differ greatly with respect to their use and mechanical properties. The mechanical

properties of adhesives are summarized in Table 1. These properties were later used to construct the triangular cohesive laws used for finite element modelling.

Adherends

The adherends materials for the studied joint configurations were chosen according to their industrial relevance. The composite used was a 0° oriented carbon-epoxy composite. CFRP is an orthotropic material, whose elastic mechanical properties can be seen in Table 2. The elastic mechanical properties of the CFRP correspond to the orientation of a 0° CFRP ply (x —fibre, y —transverse and z —thickness directions).

The metal layers used were composed of the 2024-T3 aluminium alloy. It is a high strength alloy and also has very good fatigue resistance. The mechanical properties of this alloy are shown in Table 3.

The ratio of metal and CFRP in the hybrid laminate adherend was of 1:4 (25% of metal). This ratio was selected as the proposed concept seeks the use of metal as a reinforcement of the CFRP, which ensures a minimal weight penalty. The hybrid laminate used was selected based on previous studies carried out by Carbas et al. [14]. Figure 1 shows the adherend configuration of hybrid laminates where the metal layer thickness was of 0.4 mm on each side of the CFRP core.

Specimen manufacture

As stated in the introduction, the main objective of this study was to assess the performance of hybrid adhesive joints bonded with adhesives of different stiffness. To accomplish this objective, single lap joints with an overlap of 50 mm, width of 25 mm and adhesive thickness of 0.2 mm were manufactured. The geometry of the joints is detailed in Fig. 2.

Table 1 Mechanical properties of adhesives

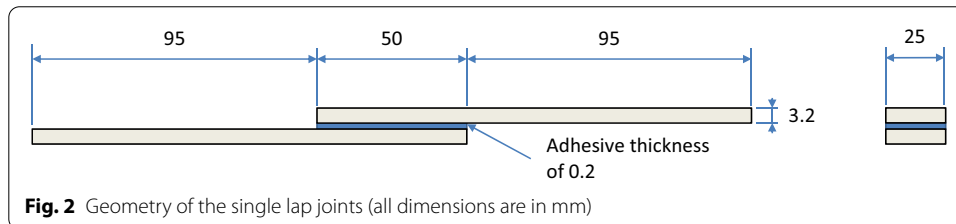
		AV138M1/HV998 [13, 14]	AF 163-2K [15, 17]	Denatite XNR 6852 E-3 [19, 20]
Quasi-static (1 mm/min)	E (MPa)	4890	1520	1728
	G (MPa)	1560	565	665
	σ_u (MPa)	39.45	46.93	51.5
	τ_u (MPa)	30.2	46.86	44.9
	G_{IC} (N/mm)	0.346	4.05	9.2
	G_{IIC} (N/mm)	4.91	9.77	51

Table 2 Elastic properties of the CFRP [21]

$E_x = 1.09\text{E}5$ MPa	$\nu_{xy} = 0.342$	$G_{xy} = 4315$ MPa
$E_y = 8819$ MPa	$\nu_{xz} = 0.342$	$G_{xz} = 4315$ MPa
$E_z = 8819$ MPa	$\nu_{yz} = 0.380$	$G_{yz} = 3200$ MPa

Table 3 Mechanical properties of 2024-T3 aluminium alclad series [22]

Young's modulus (GPa)	Yield stress (MPa)	Ultimate stress (MPa)	Poisson's ratio	Elongation (%)
66	350	440	0.3	12



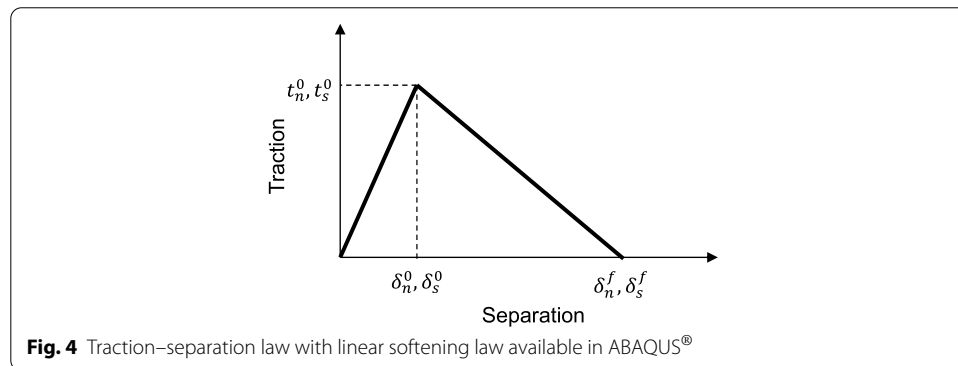
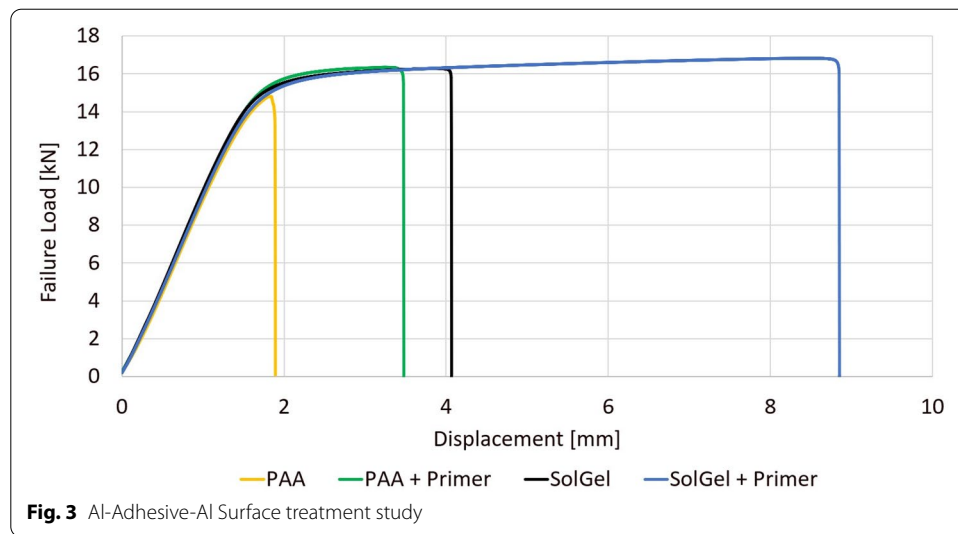
Surface treatment influence

The manufacture process of the joints required careful surface preparation of the aluminium sheets in order to ensure a good adhesion. A previous study by Carbas et al. [14], found that grit blasting and degreasing with acetone was insufficient to ensure a good level of adhesion to the aluminium laminates, resulting in a mixed cohesive-adhesive failure type for the quasi-static loading condition tested. In order to evaluate the active surface treatment that provides the best adhesion, different surface treatments were tested. To perform this analysis, SLJs bonded with AF 163 2K were manufactured and tested under quasi-static conditions. Different procedures were studied, with two different methods being considered to anodise the aluminium adherend. These two methods are phosphoric acid anodising (PAA) according to the ASTM D 3933 standard and the application of a sol-gel based anodising replacement, the 3M™ Surface Pre-Treatment AC-130-2. Moreover, the influence of a primer (a structural adhesive primer 3M Scotch Weld EW—5000 AS) was also evaluated. Load-displacement curves highlighting the relative performance of the different surface treatments can be seen in Fig. 3.

The solution that provides the best bonding results for this aluminium alloy will consequently be the one that produces the largest increase of the maximum displacement before failure. This was found to be the treatment of the surface with the sol-gel Phosphoric Acid Anodising Replacement Solution (3M™ Surface Pre-Treatment AC-130-2), followed by the application of the Structural Adhesive Primer EW—5000 AS. The application of this primer was carried out by manually brushing it unto the surface.

Tensile testing

For quasi-static conditions the single lap joints were tested using an Instron 8801 servo-hydraulic testing machine with a load cell of 100 kN at a constant crosshead speed of 1 mm/min. These tests were carried out according to ASTM D5868-01(2014) “Standard Test Method for Lap Shear Adhesion for Fiber Reinforced Plastic Bonding” [23].



Numerical analysis

Cohesive zone models are especially well suited to simulate adhesive layers, as they combine continuum mechanics (for damage initiation), and fracture mechanics (for crack propagation). Figure 4 shows the triangular cohesive zone model (CZM) used, this law is widely used due to its simplicity and provides good results for many applications [24].

FEM analyses were performed in the ABAQUS finite element software package (Dassault Systèmes Simulia Corp. Providence, RI, USA) using CZM. A bi-dimensional static analysis was used to simplify the model and reduce computational times. The elements used to mesh the model were 4-node bilinear plane strain quadrilateral elements (CPS4R). The adhesive was modelled with 4 node cohesive elements (COH2D4). Non-linear geometrical effects were included and the elastic orthotropic properties shown in Table 2 were used to simulate the behaviour of the CFRP material. The boundary conditions were consistent in all simulations and were as shown in Fig. 5. The left end of the joint was fixed while in the right end a displacement was applied to replicate the loading. The joint was also restrained transversely.

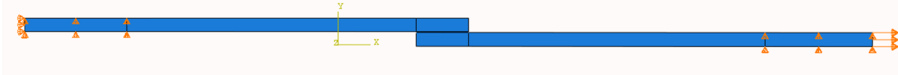


Fig. 5 Boundary conditions used

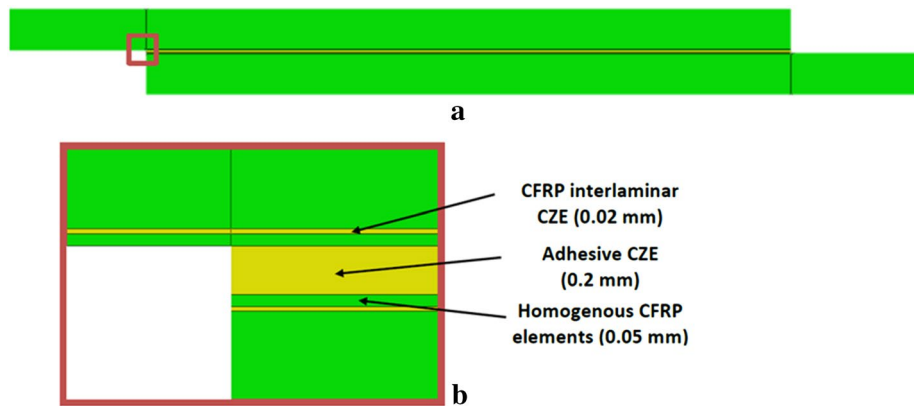


Fig. 6 **a** Detailed view of the 50 mm overlap joint; **b** enlarged detailed view in which the cohesive layers can be observed

CFRP-only joint

In order to allow for the simulation of the delamination process, a cohesive element layer (0.02 mm thick) was introduced in the adherend, at a distance of 0.05 mm from the adhesive layer. Figure 6 shows a detailed view of only-CFRP joint highlighting the location of the cohesive layers.

This model successfully replicated the results obtained experimentally. The cohesive zone elements placed between the layers of carbon fibre were found to become damaged similarly to the experimental case. Two examples of the element degradation obtained with the model are presented in Fig. 7. The stress values in the first lamina (Fig. 7a) are very high and would lead to a fibre failure.

The traction separation law is composed of a linear elastic section, which corresponds to a gradual loading until the stress criterion is achieved, followed by a softening phase, which uses fracture mechanics. The latter part of the law corresponds to the damage of the simulated material and is irreversible. In case of the law implemented in ABAQUS®, this means that if any unloading occurs, subsequent loading will not follow the initial path along the linear elastic region of the undamaged model, but one that terminates in the location where the unloading occurred. The degradation of an element is a measure of how much softening the element was subjected to.

Hybrid laminate joint

An additional model was developed in order to evaluate the behaviour of a hybrid laminate joint. This laminate uses aluminium layers with a thickness of 0.4 mm (Fig. 8). As a consequence of the high thermal stresses induced by the curing process, a two-step analysis was used, allowing to take in consideration these pre-existing stresses.

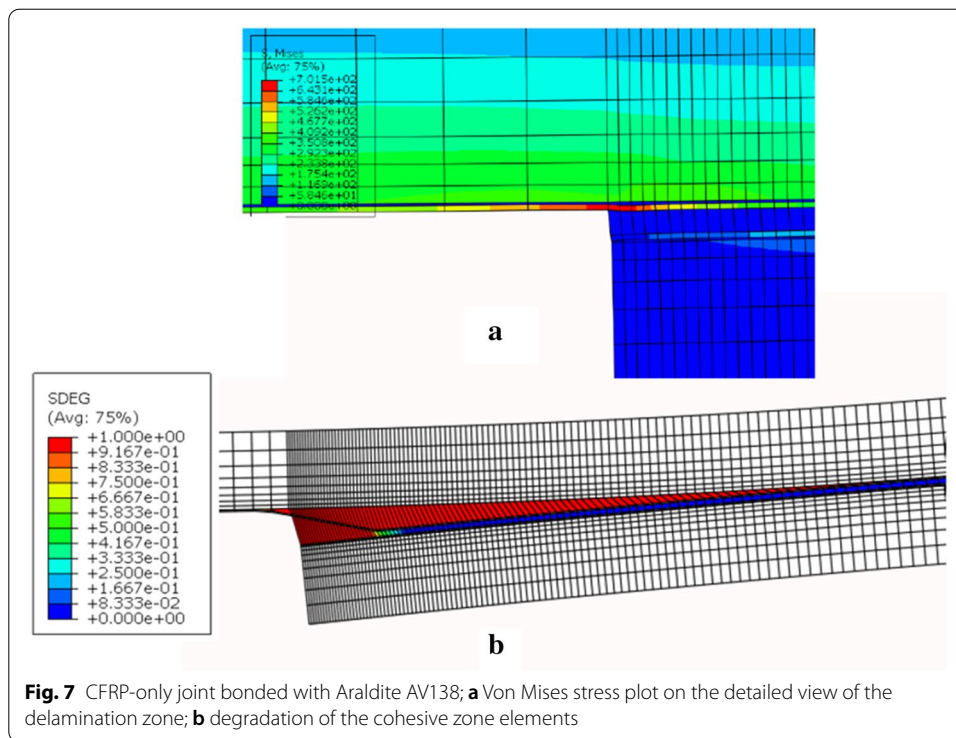


Fig. 7 CFRP-only joint bonded with Araldite AV138; **a** Von Mises stress plot on the detailed view of the delamination zone; **b** degradation of the cohesive zone elements

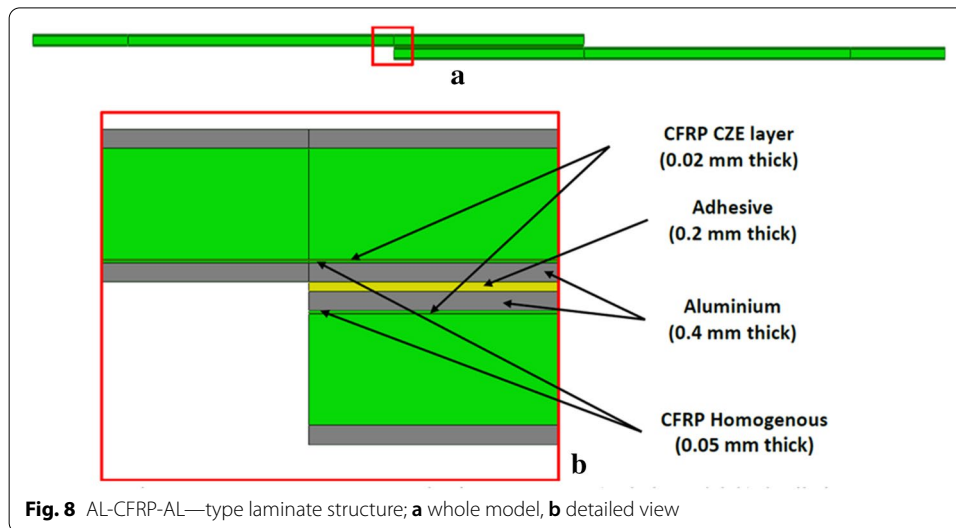
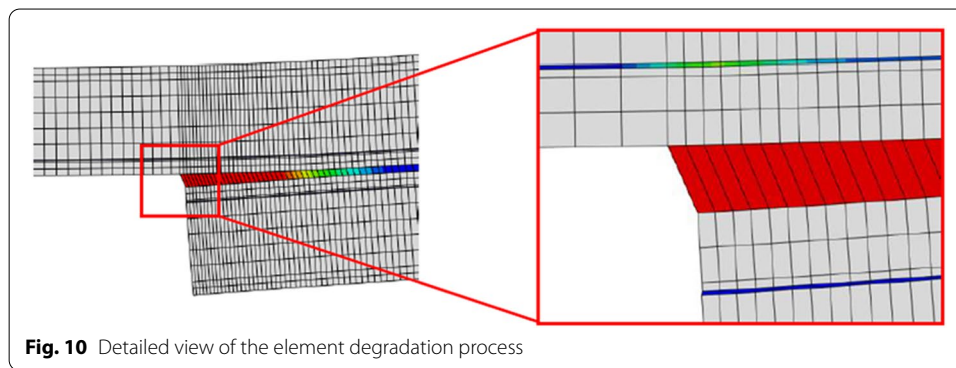
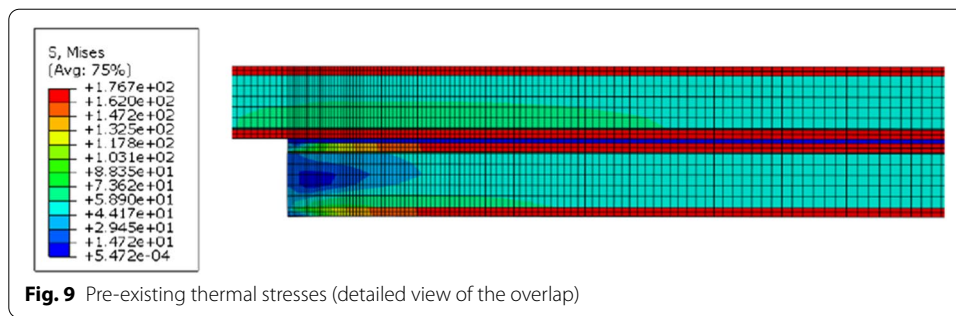


Fig. 8 AL-CFRP-AL—type laminate structure; **a** whole model, **b** detailed view

The von Mises equivalent residual thermal stresses induced due to the differences of thermal expansion coefficient between the CFRP and Aluminium is shown in Fig. 9. The contraction of the aluminium that will be present in the joint due the cure cycle of the adhesive is evident.

The isotropic aluminium layer distributes the peel stress over a larger area than a CFRP layer would do, promoting an increase of joint strength. The failure mechanism of the hybrid laminate joints bonded with AV138 is presented in Fig. 10.



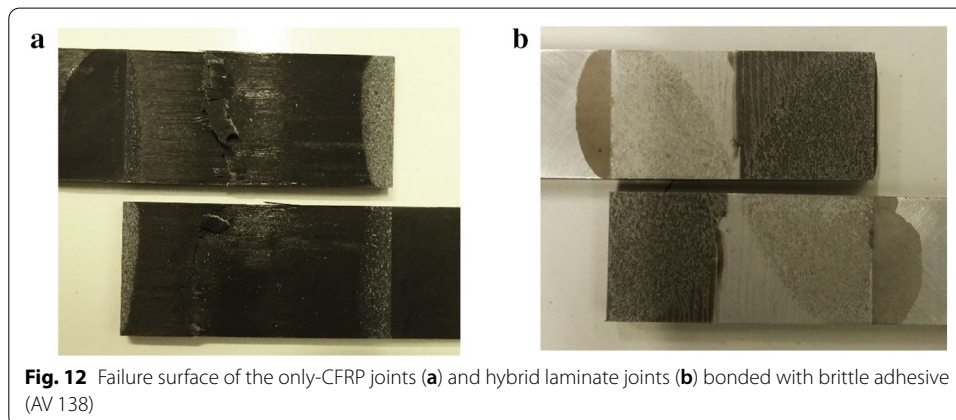
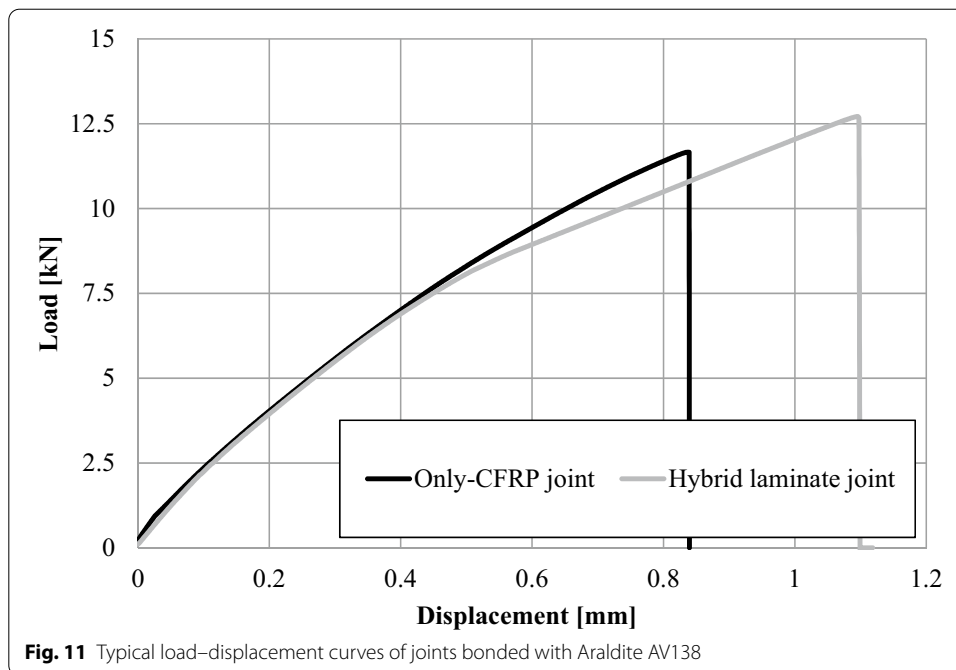
Experimental results

In order to evaluate the effectiveness of the proposed hybrid laminate joint, its mechanical performance is compared with that of joints which employ a conventional only-CFRP adherend. The test specimens were bonded with different epoxy adhesives and five specimens were manufactured for each lay-up configuration.

Araldite AV 138 joints

Figure 11 shows typical load–displacement curves for only-CFRP and hybrid laminate joints bonded with Araldite AV 138.

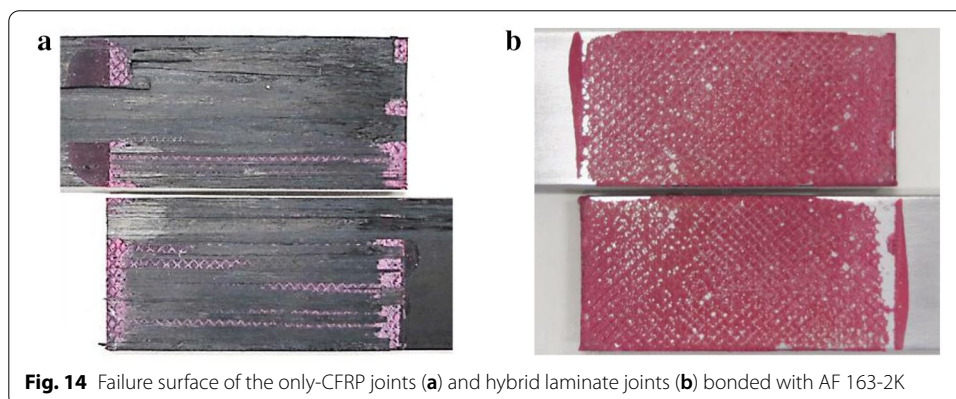
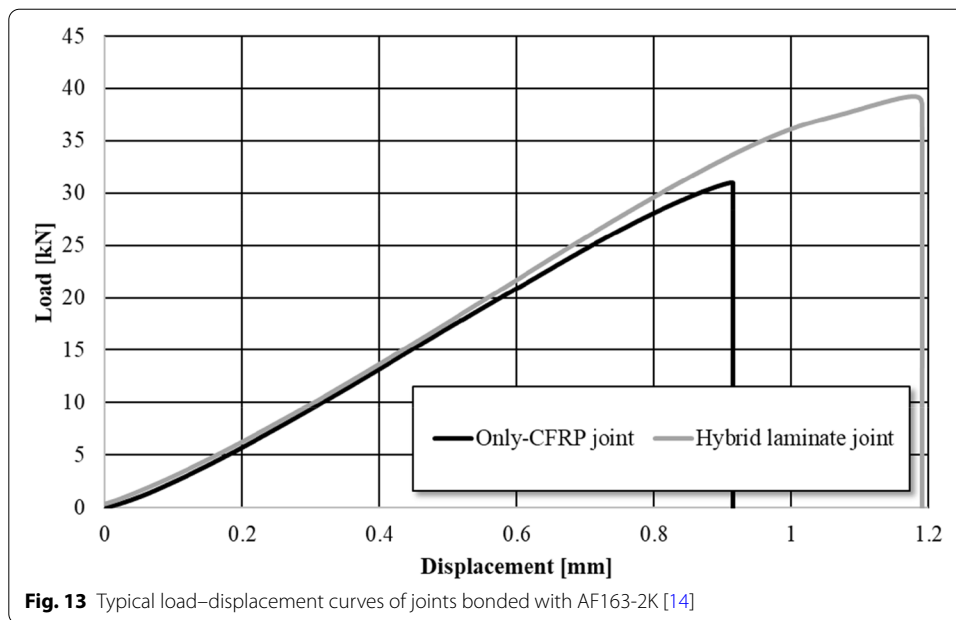
The failure load of the CFRP-only joints showed an average failure load of 11.7 ± 0.4 kN, while for the hybrid laminate joints the failure load was 12.4 ± 0.6 kN. The failure load increased marginally by 6.0% with the reinforcement of adherend with aluminium sheet (the use of hybrid laminate adherends). The failure mode for only-CFRP joints was by delamination of the adherend (Fig. 12a), while for the hybrid laminate joints a cohesive failure in the adhesive (Fig. 12b) was encountered. As expected, the external aluminium sheet led to an increase of the peel strength of the adherends and allowed for an increase of load transfer, conducting to an increase of joint strength. However, the joint strength increase is modest due to high stiffness of the adhesive, which promoted failure close to the interface where the stresses are higher. This observation suggests that a hybrid joint has the potential to provide a substantial increase of joint strength should a more ductile adhesive be used.



AF 163-2K joints

Typical load–displacement curves of only-CFRP and hybrid joints bonded with 3M AF 163-2K are shown in Fig. 13.

Carbas et al. [14] studied evaluate the joint performance of only-CFRP and hybrid laminate joints bonded with AF 163-2K. The failure load of the CFRP-only joints showed an average failure load of 31.3 ± 1.9 kN, and the hybrid laminate joints failure load reached was 39.5 ± 0.5 kN. The failure load increased by 26.2% when the adherend of adhesive joints is reinforced with an aluminium sheet. The failure mode for CFRP only joints was delamination (Fig. 14a), and for the hybrid laminate joints a cohesive failure in the adhesive (Fig. 14b) was obtained. The decrease of stiffness and increase of toughness, when compared the mechanical properties of AF 163-2K in relation to AV 138, allowed the hybrid joints to obtain a cohesive failure in the middle of bondline.



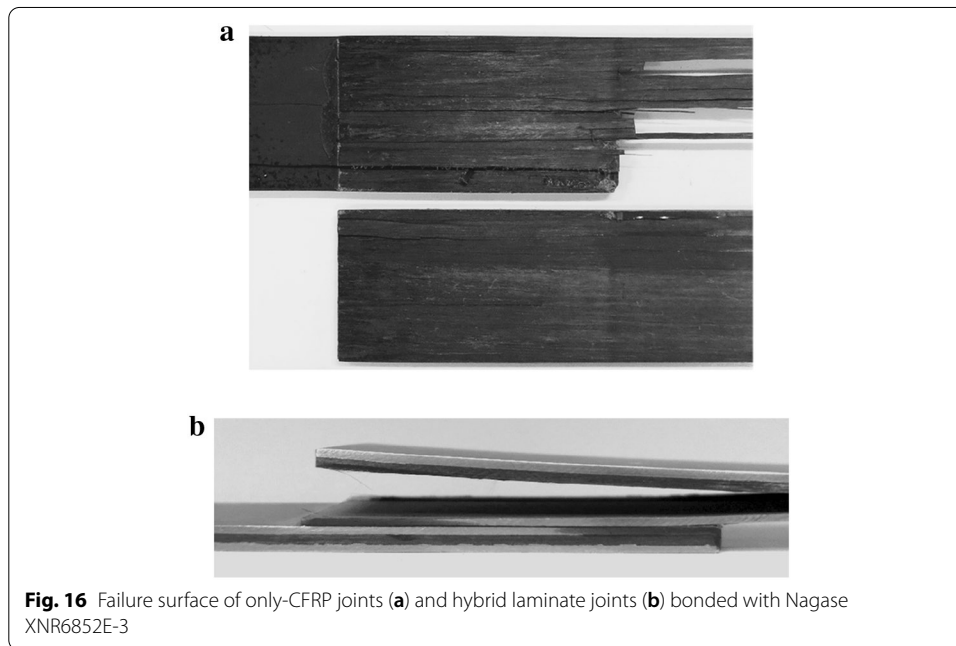
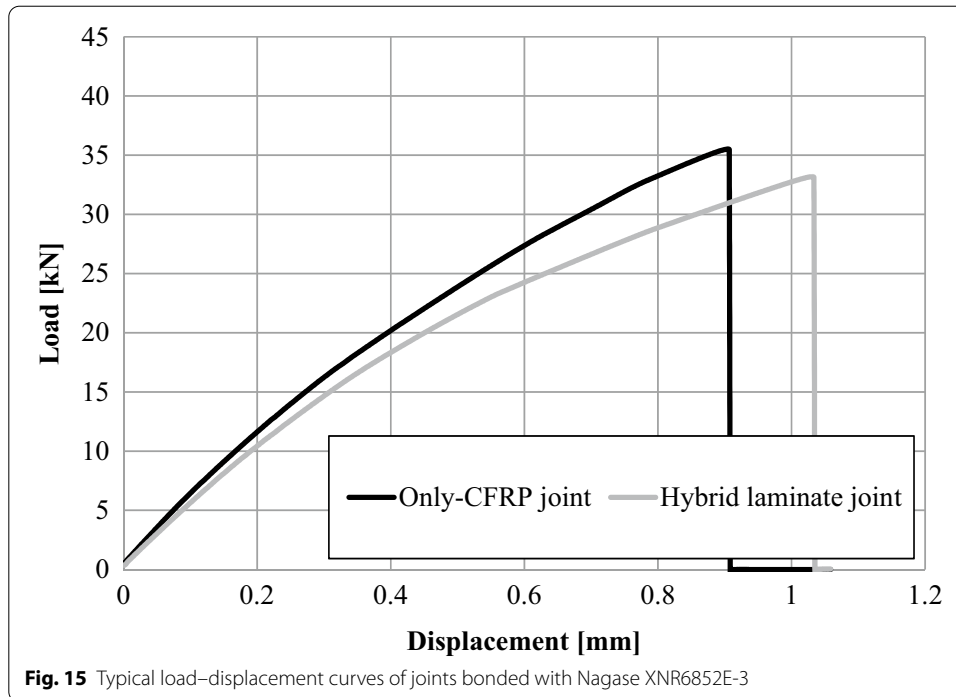
XNR6852E-3 joints

Figure 15 shows typical load–displacement curves of joints bonded with Nagase XNR6852E-3 adhesive.

The only-CFRP joints exhibited a failure load of 35.4 ± 2.2 kN while the hybrid laminate joints were found to be slightly weaker with a failure load of 32.9 ± 1.4 kN. The failure load decreased -7.0% comparing with CFRP joints. The failure mechanism for both joints bonded with Nagase XNR6852E-3 was cohesive failure in the adherend, more specifically delamination of the composite adherends (Fig. 16). This behaviour indicates that the peel strength of the adhesive is higher than the peel strength of the adherend.

Discussion of joint performance

The performance of only-CFRP joints bonded with different adhesives is shown in Fig. 17. All joints showed delamination and the adhesives with the highest toughness showed the highest strength. The high ductility of these adhesives allows them to deform



plastically and this leads to an increase in joint performance. The existence of delamination means that is theoretically possible to obtain higher joint strength if an alternative composite with higher peel strength is used.

Figure 18 shows the joint strength of hybrid laminate joints bonded with different adhesives. The AF 163-2K adhesive allows the hybrid joints to reach the highest joint strength. This indicates that both the stiffness and the strength of the adhesive should

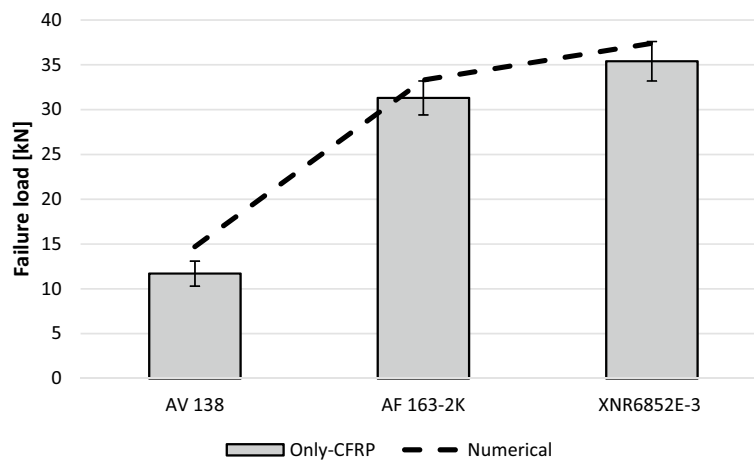


Fig. 17 Failure load of only-CFRP joints bonded with different adhesives

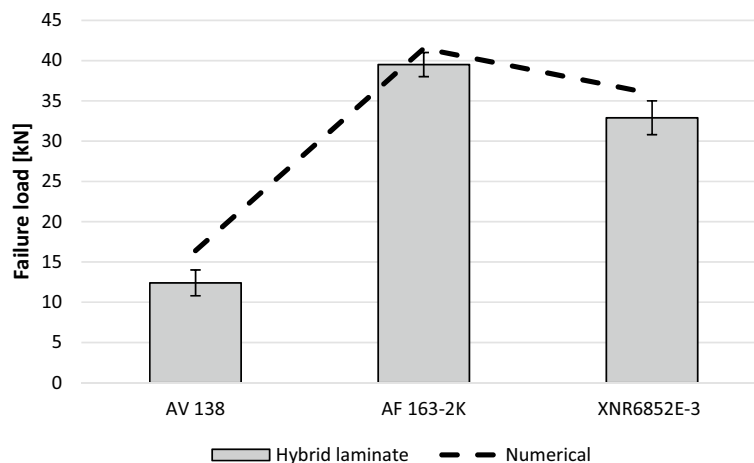


Fig. 18 Failure load of hybrid laminate joints bonded with different adhesives

may not be excessively high to attain optimal performance. In fact, the adhesive with highest toughness (XNR6852E-3) led to a delamination failure with negative consequences on joint strength.

A good correlation between the numerical models and the experimental results was achieved, not only with regards to the failure load prediction, but also in respect to failure modes and load displacement curves.

The difference in joint performance between only-CFRP and hybrid laminate joints bonded with a brittle adhesive was found to be residual at best. This is due to the stiff nature of the adhesive, introducing a high level of stress concentration at the ends of the overlap length. When an adhesive with high ductility and with high strength (Nagase XNR6852E-3) is used, the failure mechanism was found to be delamination. This is due to high peel strength of the adhesive, that promotes failure of the joints by delamination. Consequentially, the joint strength of both type of joints is the same. In contrast the adhesive with intermediate strength and with higher ductility is the adhesive that allows

to attain the largest performance improvement. This adhesive provides an effective path for load transfer through all the bonded area.

Conclusion

The main objective of this work was understanding if the strength of composite joints could be increased with the use of hybrid laminate adherends. In order to accomplish this goal, experimental and numerical procedures were carried out, considering different epoxy adhesives in order to better understand how adhesive properties influence the strength of hybrid adhesive joints. The experimental tests and numerical simulations allowed to precisely evaluate the strength and the failure mechanisms of the designed joints. This study showed that sol–gel and primer was the best surface treatment to use in order ensure a good adhesion.

Different epoxy adhesives were used, and it was demonstrated that the performance of hybrid laminate joint was heavily dependent on the mechanical properties of the adhesives used. The AF163-2K adhesive was found to behave better than stronger and stiffer adhesives in delamination prevention.

The numerical models developed in ABAQUS® showed a good agreement with the joint strength obtained experimentally. The thermal stresses analysis proved that high residual thermal stresses are present in the hybrid laminate joint, due to the very low thermal expansion coefficient of CFRP. The average equivalent von Mises stress is normally, in this setup, higher than half of the yield strength of the aluminium alloy.

Abbreviations

CFRP: Carbon fibre reinforced polymer; 3D: Tri-dimensional; *E*: Young's modulus; *ν*: Poisson's ratio; *G*: Shear modulus; PAA: Phosphoric acid anodising; CZM: Cohesive zone model.

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Authors' contributions

RJCC—This researcher carried out the experimental tests and write this part in the manuscript. EASM—This researcher carried out the numerical study and write this part in the manuscript. LFMdS—This researcher was the mentor of this concept of hybrid joints, he reviewed all the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

All relevant data is presented in the manuscript and additional information can be made available on request if necessary.

Competing interests

The authors declare that they do not have competing interests.

Author details

¹ Institute of Science and Innovation in Mechanical and Industrial Engineering (INEGI), Faculty of Engineering, University of Porto, Porto, Portugal. ² Department of Mechanical Engineering, Faculty of Engineering, University of Porto, Porto, Portugal.

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