Co-cure joining of epoxy composites with rapidly UV-irradiated PEEK and PPS composites to achieve high structural integrity

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A B S T R A C T

Efficient joining of hybrid thermoset/thermoplastic composite joints is critical to produce high performance lightweight structures while keeping the cost low. Herein, a high-power UV-irradiation technique was proposed to rapidly active the surfaces of PEEK and PPS composites for the following co-cure joining with epoxy composites. A single lap-shear joint test and a double cantilever beam test were used to evaluate the mechanical and fracture performance of the hybrid joints. The experimental results revealed that high structural integrity of the hybrid joints was achieved upon applying a 6 s UV-treatment to the thermoplastic composites. For example, the lap-shear strength and fracture energy of the adhesive bonded hybrid joints were above 25 MPa and 800 J/m², respectively. Overall, high-power UV-irradiation proved a highly efficient, rapid and low-cost method to treat thermoplastic composites for the co-cure joining with epoxy composites, and hence it demonstrated significant promise in industrial mass production.

1. Introduction

Fibre reinforced polymers (FRPs) are becoming increasingly attractive to multiple industries, including automotive and aerospace sectors, due to their lightweight and excellent structural performance. While thermosetting-based composites (TSCs) are more traditional and lower cost, thermoplastic composites (TPCs) possess a number of advantages over TSCs, including high fracture toughness, cost-effective manufacturing process and recyclability. Accordingly, TSCs, TPCs and metals with different properties are combined in various structures, that enhances design flexibility and enables the optimisation of the structures with high-performance and affordable cost. For example, the fuselage and wing sections of the new composite passenger aircrafts, Airbus A350 and Boeing 787 were manufactured from TSCs, which were connected by several thousands of TPC clips [1]. Consequently, effective joining of TPCs with TSCs and other dissimilar materials becomes critical to manufacture such components with combined materials. Three major technologies exist for the joining of thermoplastics and FRPs, including mechanical fastening [2,3], welding (fusion bonding) [4,5] and adhesive bonding [6,7]. Furthermore, an injection bonding process [8-10] has also proved to produce hybrid thermoplastic/Aluminium hybrid joints with good structural integrity, i.e. a maximum lap-shear strength of 25 MPa has been obtained in Ref. [8]. Among different joining methods, adhesive bonding offers many advantages for FRP joining, including the possibility of making lightweight constructions, the ability to join any pair of dissimilar materials with a relatively uniform stress-distribution, and the possibility to seal the entire bonding area and hence to provide high joint strength and durability. Moreover, adhesive joining is unique for joining thin-walled sections with large surface area and elements with a significant difference in thickness [11].

The majority of structural adhesives are based on thermosets, typically epoxies, who possess inherently good adhesion with the TSCs. Accordingly, the main challenge in adhesive bonding of TPC/TSC hybrid joints is to obtain good compatibility between the epoxy adhesives and the TPCs. This generally requires intensive surface treatment to the TPCs, as the majority of them possessed inherently low reactivities, small surface energies and weak polarities [12]. For example, Kinloch et al. [13-15] pointed out that a simple abrasion/solvent-wiping treatment was all that was needed to ensure a sufficiently good interfacial strength to prevent joint failure occurring at the adhesive/composite interface for the TSCs. However, in the case of TPCs, additional corona treatment was necessary to prevent the adhesive/composite interface failing at a very low value of energy. The most prevalent methods for treating TPC surfaces include acid etching [16,17], corona discharge [18,19], plasma treatment [20,21], and oxidising flame treatment [22,23]. However, many limitations exist for these methods. For example, the solutions for acid etching treatment are highly toxic.
and hence can cause severe problems for the use and disposal. The corona discharge, plasma and oxidising flame treatment methods avoid the use of wet chemicals. Nevertheless, these methods can cause non-uniformity of the treated surfaces and potential damage to the TPC surfaces. Moreover, they possess low efficiency for treating complex parts with large surfaces. Ultraviolet light (UV) irradiation proved to enhance the adhesion at different material interfaces, including copper/epoxy [24], graphene/copper [25], rubber/polyurethane [26] and silicon/gold [27]. Additionally, it is an eco-friendly and low-cost method that can uniformly treat the surfaces of the items. To date, only a limited number of studies utilised the UV-irradiation method to treat the surfaces of thermoplastics, aiming to improve the adhesion between epoxies and thermoplastics [28,29]. It was reported that the application of UV-treatment to the poly-etherether-ketone (PEEK) polymers significantly increased the lap-shear strength of epoxy adhesive bonded PEEK joints from 2.3 MPa to 13 MPa [28] and the fracture energy of PEEK film bonded TSCs from essentially zero to 820 J/m² [29]. These studies showed some potential of UV-irradiation as a surface treatment method of TPCs for their adhesively bonding with TSCs. However, little attention has been paid to this topic to date.

The aim of this work was to investigate the effects of applying high-power UV-irradiation to the TPCs on the structural integrity of co-cure bonded TPC/TSC joints. Carbon fibre reinforced epoxy prepreg were cured directly onto carbon fibre reinforced PEEK and Polyphenylene-sulphide (PPS) composites, whose surfaces were UV-irradiated for a short time of 6 s using a 200 W/cm UV bulb for a rapid surface treatment lasting for 6 s. A UV Power Puck from EIT Inc., USA was used to measure the intensities of the UV spectral ranges applied onto the TPC surfaces. The intensities of UVV (395–445 nm), UVA (320–390 nm), UVB (280–320 nm) and UVC (250–260 nm) were determined to be 1979 mW/cm², 1546 mW/cm², 343 mW/cm² and 51 mW/cm², respectively.

The carbon fibre reinforced epoxy composite (EPYC) was unidirectional prepreg, HexPly 8552-IM7-35%-134 from Hexcel. The epoxy adhesive was an aerospace-grade film adhesive, FM300M from Solvay. A layup consisting of 8 plies of carbon fibre/epoxy prepreg and one layer of film adhesive was prepared by a hand layup process, and then placed onto the consolidated TPC laminates. The assemblies were cured in a Scholz autoclave using a single dwell step at 180 °C and 4 bar gauge pressure for 90 mins, and vacuum pressure of 200 mbar inside the vacuum bag was used throughout the curing process. Hybrid joints without film adhesives between the EPYC and the TPCs were also prepared by following the same procedure. The co-cure bonding was carried out within 48 h after the application of the UV-treatment to the TPCs. The thickness of the cured EPYC was measured using an ImageJ software based on optical microscopy images of the side-view of the cured joints, as shown in Fig. 1. An average thickness of 1.02 ± 0.07 mm was obtained based on 10 measurements. It should be noted that the flexural modulus of the composite substrates was determined using a three-point bend flexural test according to ISO14125 [30], being

![Image](https://via.placeholder.com/150)

(a) Single lap-shear joint test  
(b) DCB test

Fig. 2. Schematics of (a) the single lap-shear joint test and (b) the DCB test.

2. Experimental

2.1. Materials and sample preparation

The TPCs used in this study were carbon fibre reinforced PPS composites (PPSCs) and PEEK composites (PEEKC), from TenCate Advanced Composites, the Netherlands. The 5-harness satin weave prepregs were stacked with a [0°/90°]₄s sequence and consolidated in a hot-platen press (Joos LAP100) at 2 MPa for 30 mins. The process temperature for the PPSCs and PEEKC was 320 °C and 400 °C, respectively. The thickness of the consolidated PPSC and PEEKC laminates was 2.44 ± 0.07 mm and 2.48 ± 0.09 mm, respectively. These were the average values of 10 measurements using a digital caliper (accuracy of 0.01 mm) across a 250 mm × 250 mm panel. The PPSC and PEEKC laminates were then placed in a UV-irradiation chamber equipped with a LH6 MKII UV source (200 W/cm) and a Mercury D bulb for a rapid surface treatment lasting for 6 s. A UV Power Puck from EIT Inc., USA was used to measure the intensities of the UV spectral ranges applied onto the TPC surfaces. The intensities of UVV (395–445 nm), UVA (320–390 nm), UVB (280–320 nm) and UVC (250–260 nm) were determined to be 1979 mW/cm², 1546 mW/cm², 343 mW/cm² and 51 mW/cm², respectively.

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![Image](https://via.placeholder.com/150)

(a) Hybrid joint without adhesive  
(b) Adhesive bonded hybrid joint

Fig. 1. Representative side views of the TPC/TSC joints without and with a layer of film adhesive in between.
162 GPa, 68 GPa and 65 GPa for the EPYC, PPSC and PEEKC laminates, respectively. Accordingly, the configurations of the hybrid joints (see Fig. 1) were defined to ensure an essentially pure mode-I fracture mode in the following double cantilever beam (DCB) tests. After the curing, specimens with desired dimensions were cut out from the cured joints for the following tests. It should be noted that all the hybrid joints consisting of non-treated TPCs failed during the cutting process. This was due to the typically poor adhesion at the epoxy/PPS and epoxy/PEEK interfaces.

2.2. Testing and analysis

The chemical composition of the non-treated and UV-treated surfaces of the TPCs were analysed using a X-ray photoelectron spectrometer (XPS, Kratos Axis Ultra DLD), equipped with an Al Kα (1486.7 eV) X-ray source. A mobile surface analyser from KRÜSS, GmbH was used to determine the surface free energies and water contact angles of TPC surfaces. The tensile lap-shear strength of the TPC/TSC joints was measured using a single lap-shear joint test, as schematically shown in Fig. 2(a). The lap-shear test was carried out using a universal testing machine (Zwick-Roell Z250 SW) at a constant loading rate of 2 mm/min at ambient temperature. The specimens were fixed to the machine using a pair of hydraulic clamps with a clamping pressure of 200 bar. A misalignment between the upper and lower clamps was set to ensure the applied force was in the middle plane of the overlap during the test. The fracture behaviour of the hybrid joints was investigated using a mode-I DCB test according to ASTM D5528, see Fig. 2(b). The test was carried out using a Zwick/Roell ZO10 testing machine at a constant displacement rate of 2 mm/min at ambient temperature. Three replicate tests were conducted in each case for both of the single lap-shear joint test and the DCB test. To analyse the failure mechanisms of the joints, the fracture surfaces of the tested specimens were imaged using a VK-X1000 microscope from KEYENCE Corporation.

3. Results and discussion

3.1. Surface characterisation

Table 1 shows the results of the surface characterisation of the non-treated and UV-treated TPCs. It was observed that applying the UV-treatment to the TPCs notably increased the O/C ratios of the TPC surfaces in both cases. This was achieved by the breakage of the C=C/C=H bonds upon the UV-irradiation and the generation of C=O, C=O and O-C=O species [15,28,31,32]. Consequently, the presence of more oxygen functional groups on the TPC surfaces significantly increased their polar surface energies ($\gamma^p$), and subsequently increased the ratios

![Fig. 3. Illustration of the interactions at the interface of two phases [33].](image3)

![Fig. 4. Water contact angles of the non-treated and UV-treated PPSC and PEEKC laminates.](image4)

![Fig. 5. Typical microscopy images of the surfaces of the EPYC, PPSC and PEEKC laminates.](image5)
between $\gamma_p$ and the dispersive surface energies ($\gamma_d$). As shown in Table 1, the $\gamma_p/\gamma_d$ ratio increased from 0.03 of the non-treated PPSC to 0.09 of the UV-treated PPSC, and from 0.08 of the non-treated PEKKC to 0.12 of the UV-treated PEEKC. It is worthy noting that only the same types of forces, i.e. dispersive-dispersive, or polar-polar, interacted at the interface of two components, as shown in Fig. 3. Clearly, a better match of the $\gamma_p/\gamma_d$ ratios of the two phases is desirable to achieve good interactions at their interface. The values of $\gamma_p$ and $\gamma_d$ of epoxy adhesives were reported by Kinloch [34] to be about 5–8 mN/m and 40 mN/m, respectively. This corresponded to a $\gamma_d/\gamma_p$ ratio of between 0.125–0.2. Obviously, the application of the UV-treatment to the TPC surfaces resulted in a closer match of the $\gamma_p/\gamma_d$ ratios at the adhesive/TPC interfaces in this work. This subsequently decreased the water contact angles of the TPC surfaces from 83.47° to 71.86° for the PPSC substrate, and from 80.22° to 67.77° of the PEEKC substrate, as shown in Fig. 4. Overall, the surface characterisations of the TPCs proved UV-irradiation a highly effective method for surface activation of the PPSC and PEEKC laminates. This significantly enhanced the structural integrity of the hybrid joints, as will be shown in Sections 3.2 and 3.3.

Fig. 6. The lap-shear strengths of the UV-treated hybrid joints.

Fig. 7. Typical images of the fracture surfaces of the lap-shear specimens of the UV-treated joints: (a) without film adhesives and (b) with film adhesives.
As noted in Section 2.1, the hybrid joints failed prior to test if the TPC surfaces were not treated by the UV-irradiation. This clearly indicates a very poor adhesion at the epoxy/non-treated TPCs interfaces.

Fig. 5 shows typical microscopy images of the surfaces of the EPYC, PPSC and PEEKC after the failure of the non-treated joints. As expected, a clear surface without any evidence of damage was observed in all cases. It is worthy to note that Fig. 5 also represents typical surface morphologies of the non-damaged laminates, which can be used as references to be compared with the fracture surfaces of the single lap-shear joint test and the DCB test specimens presented in Sections 3.2 and 3.3.

3.2. Single lap-shear test

The lap-shear strengths (LSSs) of the hybrid TPC/TSC joints from the single lap-shear joint tests are summarised in Fig. 6. In Fig. 6, EPYC/PPSC(UV) means the joints between the EPYC and the UV-treated PPSC without a layer of adhesive in between, and EPYC/A/PPSC(UV) indicates the joints bonded with adhesives. It should be noted that the LSSs were not measurable for the non-treated hybrid joints as they failed prior to test in all cases. It is clear that the application of 6 s UV-treatment to the surfaces of the PEEKC and PPSC laminates significantly increased the LSSs of the hybrid joints. In particular, the LSSs of the hybrid joints increased from essentially zero to 20.7 MPa and 17.2 MPa for the EPYC/PPSC(UV) joints and the EPYC/PEEKC(UV) joints, respectively. More prominent increases in the LSSs were achieved for the adhesive bonded joints upon the UV-treatment, i.e. a LSS of 25.2 MPa and 25.4 MPa was observed for the EPYC/A/PPSC(UV) joints and the EPYC/A/PEEKC(UV) joints, respectively.

The fracture surfaces of the lap-shear specimens were analysed to correlate the significantly enhanced lap-shear strength with the failure locus and mechanisms of the hybrid joints. Fig. 7 shows typical images of the fracture surfaces of the lap-shear specimens joined without and with film adhesives. For the hybrid joints without film adhesives, a large amount of white colour PPS or PEEK polymers appeared on the surfaces of the UV-treated PPSC and PEEKC substrates, leaving bare carbon fibres on the corresponding location of the opposite TPC sides, as shown in Fig. 7(a). This indicates a substrate damage to the TPCs during the lap-shear test, i.e. the thermoplastic polymers on the surfaces of the UV-treated PPSC and PEEKC substrates were damaged and peeled-off from the carbon fibres. An alternation of a cohesive failure in the adhesive layer and a substrate damage to the TPC substrates was observed for the hybrid joints bonded with adhesives, as shown in Fig. 7(b). The damage to the TPCs was more severe when compared to their counterparts without film adhesives. i.e. apart from damage and debonding of the thermoplastic polymers, extensive carbon fibre delamination and breakage also took place. All these observations demonstrated that applying the UV-treatment to the TPCs significantly improved the adhesion with epoxies to a level that was sufficient to cause substrate damage to the PEEKC and PPSC substrates during the lap-shear test. This resulted in remarkable increases in the LSSs of the adhesive joints, i.e. from essentially zero to a value of between 17.2 MPa and 25.4 MPa, as shown in Fig. 6.

3.3. Mode-I fracture test

The load versus displacement curves and corresponding R-curves from the DCB tests of the UV-treated hybrid joints are shown in Fig. 8. A stick-slip fracture behaviour took place for the PEEKC(UV)/EPYC joints, evidenced by the zigzag shape of the corresponding load versus displacement curve in Fig. 8(a). In this case, the values of the load at the peak points were used for the calculation of the fracture energy. Apart from the PEEKC(UV)/EPYC joints, all the other hybrid joints exhibited a stable fracture propagation manner. It is clear that the failure loads of the DCB specimens were much higher for the joints bonded with adhesives than their counterparts without adhesives. This corresponded to higher fracture energies of the adhesive bonded joints, as shown in Fig. 8(b). Moreover, the R-curves of the adhesive
bonded joints exhibited an obviously ‘rising’ trend for both of the PPSC (UV)/A/EPYC and PEEKC(UV)/A/EPYC joints, indicating the presence of extensive fibre bridging during the fracture process. The mode-I fracture propagation energies, $G_{IC}$ of the hybrid TPC/TSC joints are summarised in Fig. 9. The fracture energies of the hybrid joints without adhesives were measured to be 335 J/m$^2$ and 316 J/m$^2$ for the PPSC(UV)/EPYC and PEEKC(UV)/EPYC joints, respectively. These values were slightly lower than the interlaminar fracture energy of the EPYC, that was reported to be 380 J/m$^2$ in Ref. [29]. The fracture energies of the UV-treated PPSC(UV)/A/EPYC and PEEKC(UV)/A/EPYC joints were more or less the same, i.e. being 885 J/m$^2$ and 836 J/m$^2$, respectively.

An analysis on the fracture surfaces of the DCB specimens was carried out to understand the energy dissipation mechanisms during the fracture process. Typical images of the fracture surfaces of the DCB specimens bonded without and with adhesives are shown in Fig. 10. Some damage to the TPC substrates took place for the hybrid joints without adhesives, evidenced by the presence of many spots of white colour PEEK or PPS polymers on the EPYC sides and signs of polymer damage on the TPC surfaces, see Fig. 10(a). While this phenomenon indicates relatively good adhesion at the EPYC/TPC interfaces, the lack of resin at the fracture plane resulted in the relatively low mode-I fracture energies, as shown in Fig. 9. For the adhesive bonded joints, the majority of the adhesive together with a large amount of broken carbon fibres were observed to be well-attached to the TPC sides, leaving a small amount of adhesive on the severely damaged EPYC substrates. The presence of the delaminated and broken carbon fibres on the TPC surfaces, together with the ‘rising’ trend of the $R$-curves in Fig. 8(b) typically indicate extensive carbon fibre bridging during the fracture process. Overall, the significantly enhanced adhesive/TPC interface adhesion upon the UV-treatment resulted in severe damage to the adhesive layers and the EPYC substrates during the fracture process, and subsequently resulted in the high fracture energies of the adhesive bonded hybrid joints, as shown in Fig. 9.

3.4. Discussion

The experimental results of this work demonstrated that applying a rapid UV-treatment to the TPCs significantly enhanced the structural integrity of the TSC/TPC hybrid joints bonded with and without adhe-
Furthermore, the use of adhesives was required to achieve the best performance of the hybrid joints, especially for the fracture behaviour. For the adhesive bonded joints, applying a rapid UV-treatment to the TPCs successfully transformed the failure mode of the hybrid joints from pure interfacial failure (with a bonding strength of essentially zero) to substrate damage and cohesive failure in the adhesive. The absence of interfacial failure typically indicated that the adhesive joints obtained the highest structural integrity that can be achieved by using a surface treatment strategy. Accordingly, the high-power UV-irradiation technique had proved comparable efficiency for adhesion improvement as plasma treatment, which is currently the most effective and industrially approved process for activating thermoplastic surfaces. For example, it was reported that applying atmospheric-pressure plasma (APP) [35–38] and low-pressure plasma (LPP) [39–42] treatments to the PEEK and PPS surfaces also resulted in substrate damage and/or cohesive failure of the adhesive joints during the lap-shear test. However, there are a number of limitations of plasma treatments for industrial applications. For instance, the parts have to be placed in a vacuum chamber for the LPP treatment, that resulted in a number of disadvantages, including the high processing cost, the difficulties to integrate the treatment process into existing process chains and the infeasibility to treat large parts [43]. For these reasons, the use of LPP is often not applicable in industrial applications. The development of APP process had successfully avoided these shortcomings of the LPP treatment. Nevertheless, the processing temperature at the nozzle outlet can be very high for the APP treatment, which can easily cause damage to the thermoplastic surfaces, e.g. through melting [44]. Moreover, the size of the plasma nozzle is relatively small. This can negatively affect the time efficiency and treatment uniformity of the APP process while treating large parts and complex surfaces. Accordingly, apart from the high efficiency for adhesion improvement, the proposed high-power UV-irradiation process possessed a number of advantages for industrial applications, such as the rapid treatment, the relatively low cost, the feasibility to treat large parts with complex surfaces and the possibility to achieve uniform treatment and to be employed directly in the production line (e.g. a UV-source can be easily installed above the belt of a conveyor system). Consequently, it showed significant promise to replace plasma treatments for the surface activation of thermoplastics and TPCs for adhesive bonding.

4. Conclusions

A high-power UV-irradiation technique was used to rapidly activate the surfaces of carbon fibre reinforced PPS composites (PPScs) and PEEK composites (PEEkcks) for a short time of 6 s. This notably increased the amount of oxygen element on the composite surfaces and decreased their water contact angles. A layup of epoxy composite (EPYc) prepregs was then cured directly onto the PPSc and PEEkc panels, without and with a layer of film adhesive in between, to produce hybrid thermoset/thermoplastic composite joints. All the hybrid joints consisting of non-treated PPSc and PEEkc substrates failed prior to test, as a result of the typically poor adhesion between epoxies and thermoplastics. Encouragingly, the application of a 6 s UV-treatment to the PPSc and PEEkc substrates significantly improved their adhesion with epoxies, that remarkably increased the lap-shear strength and fracture energy of the hybrid joints in all cases. For example, a lap-shear strength of above 25 MPa and a mode-I fracture energy of 800 J/m2 were observed for both the adhesively bonded PPSc/EPYc and PEEkc/EPYc joints. The results of this work demonstrated significant promise of high-power UV-irradiation as a rapid, effective and low-cost method for the surface preparation of thermoplastic composites, regarding mass production of co-cured PPSc/EPYc and PEEkc/EPYc joints with high structural integrity.

Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time due to legal or ethical reasons.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Dong Quan: Conceptualization, Investigation, Writing - original draft, Funding acquisition. René Alderliesten: Funding acquisition, Project administration, Writing - review & editing. Clemens Transfeld: Resources, Methodology, Writing - review & editing. Ioannis Taskoniatis: Resources, Investigation. Rinze Benedictus: Resources, Funding acquisition.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.composites.2020.112595.

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