

# Bond Strength of Co-Bonded Thermoplastic Leading Edge Protection (LEP): The Effect of Processing-Driven Interphase Morphology

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**Abstract.** Integrated leading edge protection (InLEP) is a novel LEP method that involves co-bonding a tough thermoplastic to the blade shell of the wind turbine made of fiber-reinforced thermoset polymer. In the co-bonding process, as a result of the interdiffusion of the bonded thermoplastic and thermoset polymers, an interphase is formed between them. An important factor affecting the level of interdiffusion is the cure temperature. In this work, we investigate the influence of cure temperature on the interphase morphology and bond strength of ABS-polyester/glass and PC-polyester/glass hybrid composites. The hybrid composites are manufactured via vacuum assisted resin transfer molding. Interphase morphology is observed and the interphase thickness is measured via optical microscopy. Bond strength is tested via climbing drum peel testing and subsequently, fractography analysis is carried out on the fractured samples. It was found that both the interphase thickness and bond strength decrease with an increase of cure temperature. The decrease in bond strength at high temperatures was accompanied by an increase in the extent of interfacial failure, while interphase failure at low temperatures promoted higher bond strength.

## Introduction

Leading edge erosion is a prominent problem for offshore wind turbine blades due to extremely high tip speeds and harsh climate conditions [1]. The wind industry strives to mitigate this problem by developing leading edge protection (LEP) systems. Common LEP solutions suffer from insufficient impact resistance of the LEP material, compromise of the aerodynamic performance due to the change of blade shape after the LEP application, or failure at the interface between the LEP and the main blade shell as they are adhesively bonded [2]. A novel LEP solution called the integrated LEP (InLEP) [3], which does not alter the aerodynamic shape of the blade or require adhesives, involves using a tough thermoplastic (TP) polymer as the LEP material co-bonded to the wind turbine blade shell. Herein, as-manufactured TP is first laid on the mold together with the dry fabrics of the blade, and subsequently, the dry fabric is infused with thermoset (TS) resin in a vacuum assisted resin transfer molding (VaRTM) process. Driven by the thermodynamic affinity between the TS resin and TP polymer, the TP polymer swells and the TS resin diffuses into the TP polymer; hence, interdiffusion takes place between the two polymers, resulting in the formation of an *interphase*. Upon the completion of cure, a bond is formed without the use of any adhesives. The thickness and morphology of the interphase were shown to be highly dependent on the thermodynamic affinity, cure, and diffusion kinetics of the TS resin and the physical state of the TP [4–8]. For instance, a lower viscosity and a higher gelation time of the resin both promote a higher interphase thickness [6,7]. This is because a lower viscosity results in enhanced mobility of the polymer molecules and a higher gelation time provides more time for the molecules to diffuse.

Since the interphase is the physical link between the bonded polymers, it is of utmost importance to investigate its effect on the bond strength and optimize its thickness/morphology to have the optimum bond strength. In two former studies [6,7], acrylonitrile butadiene styrene (ABS) and polycarbonate (PC) were found to be highly compatible with the unsaturated polyester resin, which is used for manufacturing wind turbine blades, forming interphases with high thickness. Later, we investigated the effect of the interphase thickness and morphology of these materials systems on the

bond strength in a recent study [9]. We found that ABS formed a homogeneously mixed interphase with polyester/glass, while phase separation took place in PC-polyester/glass. Furthermore, ABS-polyester/glass had a softer interphase with a higher thickness [9]. Eventually, the bond strength of ABS-polyester/glass was significantly higher, which was attributed to the extensive interphase (and intra-ply) failure, while the failure of PC-polyester/glass was mostly interfacial (between the interphase and polyester/glass). Hence, a homogeneously mixed and soft interphase was seen to favor a higher bond strength. Although we compared different material systems in that study [9], the effect of processing conditions, or specifically the cure temperature, was not investigated. Cure temperature is known to affect the interdiffusion process since both the viscosity and gelation time, which influence the interphase thickness and morphology, depend on temperature [6,7].

In this study, we aim to determine the effect of cure temperature on the interphase morphology and bond strength of the co-bonded ABS-polyester/glass and PC-polyester/glass hybrid composites. For this purpose, the hybrid composites are manufactured via VaRTM at different cure temperatures. The interphase morphology is characterized and the interphase thickness is measured by optical microscopy. The bond strength of the hybrid composites is measured via Climbing Drum Peel (CDP) test. The fractured specimens are subjected to fractography using an optical microscope. Finally, interphase morphology, bond strength, and fracture behavior are correlated.

## Experimental

**Materials.** As the matrix of the fiber-reinforced thermoset (FRT) layers, unsaturated polyester resin (UPR) with 45 wt% styrene suitable for wind turbine blade manufacturing was used. The resin was cured with a liquid peroxide initiator. The ratio of the weight of the peroxide to that of UPR was 1.5%. As TP materials, ABS and PC plates with grade names VIKUREEN ABS PLAAT GLANS WIT 0291 and LEXANTM 9030, respectively, were used. The thickness of the ABS and PC plates were 3 mm and 2 mm, respectively. Fiber reinforcement of the FRT layers was a glass fabric with a total areal weight of 750 g/m<sup>2</sup> composed of unidirectional (UD) roving (660 g/m<sup>2</sup>) and stitching and randomly oriented transverse fiber bundles on one side of the fabric (90 g/m<sup>2</sup>). The fiber architecture of the fabric is presented in Figure 1.

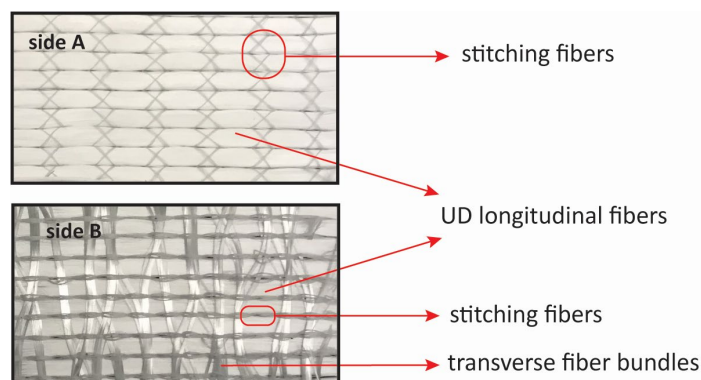


Figure 1: Two sides of the fabric used as reinforcement.

**Laminate manufacturing.** The hybrid composites, which consist of a thermoplastic plate sandwiched between one layer of glass/polyester on each side, were manufactured via VaRTM (schematically illustrated in Figure 2). The sandwich stacking was chosen to ensure a balanced lay-up that prevents the curing-induced warpage of the laminate. In the stacking, the random transverse fibers were in contact with the thermoplastic plate and the longitudinal 0° fibers were along the crack propagation direction, as shown in Figure 3. To create pre-crack for peel tests, a polyimide film was placed between the fabric on one side and the thermoplastic plate. After mixing the resin with the initiator for 3 minutes and degassing for about 5 minutes, the mixture was infused at room temperature through the fabric by applying full vacuum pressure. Subsequently, the fully vacuumed stack was cured for 24 hours at the initial cure temperature in a preheated oven. To investigate the effect of cure

temperature on interphase morphology and bond strength, different initial cure temperatures were applied (25°C, 35°C, and 50°C for ABS-polyester/glass; and 25°C and 50°C for PC-polyester/glass). Afterward, the stack was kept in an oven together with the mold for postcure at 60°C for 24 hours.

The cured ABS-polyester/glass and PC-polyester/glass laminates had thicknesses of approximately 4.8 and 3.8 mm, respectively. Subsequently, specimens for peel tests with a width of 10 mm and length of 265 mm were cut from the laminates (pre-crack length: 95mm). The fiber volume fraction of the polyester/glass layers was measured to be 55% through burn-off tests.

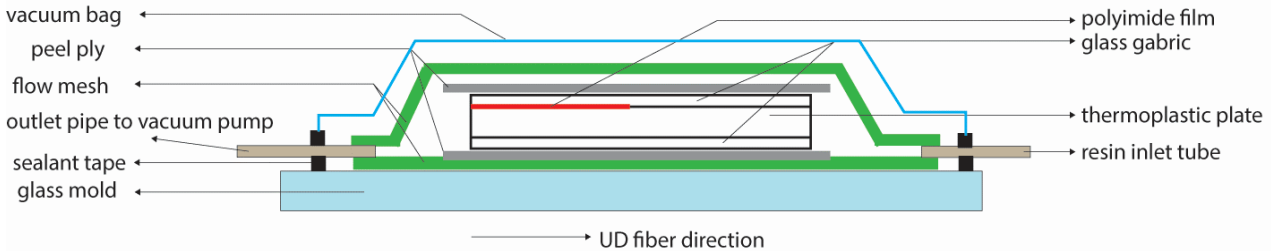


Figure 2: Schematic illustration of the VaRTM setup.

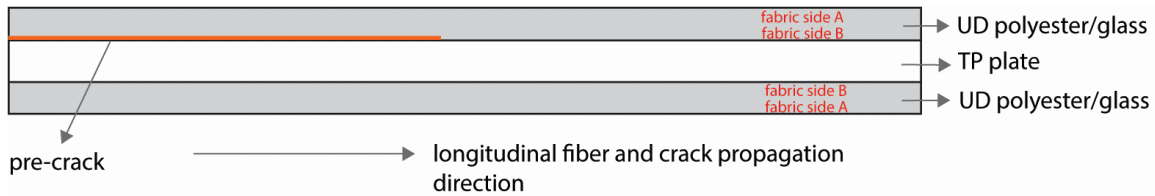


Figure 3: Schematic drawing of the lay-up and the peel test specimen.

**Interphase morphology.** The interphase morphology of the hybrid composites was observed and the interphase thickness was measured by optical microscopy analysis. At least three cross-sections of dimensions 18mm x 18mm were cut from each laminate cured at different temperatures. These cross-sections were later polished using a Struers Tegramin 30 polisher. Micrographs were made using a Keyence VHX-7000 optical microscope. Since the interphase thickness varies throughout the laminate as a result of heterogeneous fiber distribution, the interphase thickness corresponding to each cross-section was determined by averaging the interphase thickness along the length of the cross-section. This was done by dividing the area of the interphase by the length of the cross-section, as explained in [9].

**Fracture tests and fractography.** Bond strength was evaluated by Climbing Drum Peel (CDP) tests. A CDP apparatus based on ASTM D1781 [10] was used. A crosshead speed of 20 mm/min was applied using a Zwick 1445 universal testing machine equipped with a 1 kN load cell. First, the tests were run with the uncracked specimens and the total force ( $F_d$ ) resulting from the crack propagation and the weight of the drum was recorded. Subsequently, as proposed in [11], the cracked specimens were also tested in the same range of crosshead positions as the uncracked specimens to determine the force response due to the weight of the drum ( $F_w$ ). Peel energy  $G_{IC}$  was calculated by making use of the net force  $F_d - F_w$ , as suggested in [12]:

$$G_{IC} = \frac{F_d - F_w}{w} \cdot \frac{r_2 - r_1}{r_1}, \quad (1)$$

where  $w$  is the width of the specimen,  $r_1$  is the radius of the drum including one-half of the thickness of the peeled skin (polyester/glass layer), and  $r_2$  is the radius of the flange including one-half of the thickness of the loading straps.  $r_1$  and  $r_2$  in this study are 50.5 and 65.25 mm, respectively. Five specimens were tested per processing temperature for each material system.

After peel tests, fractured samples were subjected to fractography by making use of optical microscopy. To visualize the crack path, the peeled skin and the base laminate were clamped together

and embedded in Epofix resin. The embedded samples were later polished using a Struers Tegramin 30 polisher and observed with Keyence VHX-7000 optical microscope.

## Results

**Interphase morphology.** Figure 4 shows the optical micrographs of ABS-polyester/glass and PC-polyester/glass hybrid composites manufactured at various cure temperatures. In the micrographs, different phases (thermoplastic, interphase, and the thermoset composite) can clearly be identified. This shows the suitability of optical microscopy analysis to observe interphase morphology, specifically the interphase thickness.

Interphase of the hybrid composite with ABS shows a fine, homogeneous-looking morphology at each cure temperature. Homogeneous mixing was confirmed for the sample cured at room temperature by the almost constant modulus and hardness across the interphase in our previous study [9]. The dark regions at the boundaries of the interphase are due to the polishing procedure, where some locations in the soft interphase undergo more material removal during polishing; hence they appear dark. Unlike ABS-polyester/glass, PC-polyester/glass exhibits two different regions: the phase-separated region close to polyester/glass and the dark, homogeneously mixed region close to PC (for a detailed discussion of the interphase morphology, see [6,9]). The micrographs at different temperatures show that the interphase morphology is similar at different cure temperatures for both hybrid composites. The observation that the phase separation takes place at the interphase of polyester/glass with PC but not with ABS can be explained by the lower thermodynamic affinity between PC and UPR [6,7,9,13].

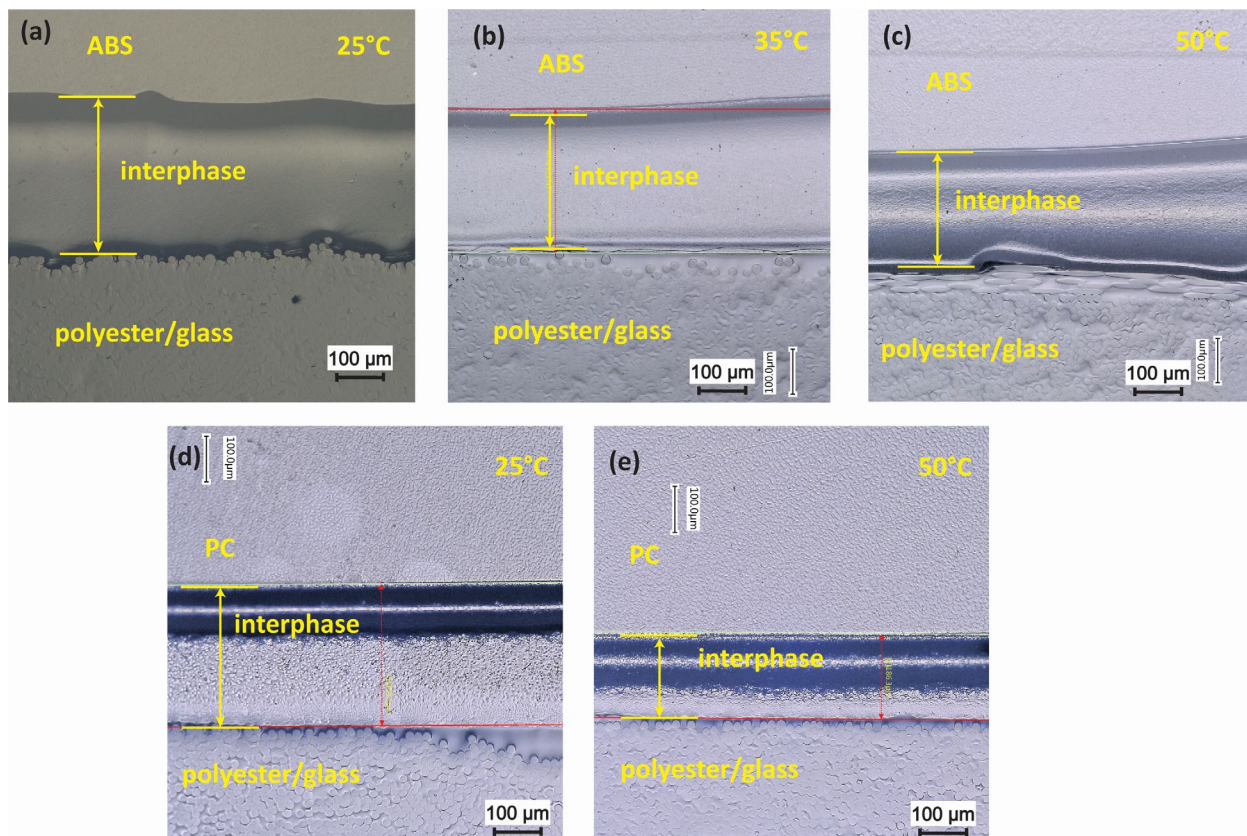


Figure 4: Optical micrographs of ABS-polyester/glass cured at (a) 25°C, (b) 35°C, (c) 50°C and PC-polyester/glass cured at (d) 25°C, (e) 50°C.

Next, the interphase thickness of the hybrid composites calculated by the method mentioned in the “Experimental” section and its dependence on the cure temperature is discussed. Comparing the interphase thickness of ABS-polyester/glass and PC-polyester/glass shown in Figure 5a, it can be seen that the use of ABS in the hybrid composite leads to overall a higher interphase thickness. This



trend agrees with its higher affinity with polyester. Although changing the cure temperature does not influence the interphase morphology as seen in Figure 4, it leads to considerably different interphase thicknesses. An increase in the cure temperature leads to a decrease in the interphase thickness for both hybrid composites. This might indicate that the interphase thickness of the hybrid composites is controlled by the gelation time. Since an increase in cure temperature lowers the gelation time, less time is available for the interdiffusion to take place, which leads to a decrease in the interphase thickness. The observed trend is different from that observed for the neat polymers [6,7] shown in Figure 5b, where the interphase thickness first decreases from 25°C to 35°C, but shows an increase from 35°C to 50°C. In previous research [6,7], this was attributed to competing temperature dependence of the gelation time and viscosity of the resin. From 35°C to 50°C, a sharp decrease of viscosity of the resin compared to a more moderate decrease of gelation time led to the effect of viscosity being more dominant in the resulting interphase thickness. Hence, our contradicting observation for the hybrid composites shows that the dependence of gelation time on cure temperature may be more dominant in the interphase thickness than that of gelation time. Nevertheless, to clarify this, the effect of fiber reinforcement on interdiffusion should be studied in more detail in future work.

Comparing the interphase morphology seen in Figure 4 to that between the neat polyester and ABS and PC thermoplastics from the works of Zanjani and Baran [7] and Zanjani et al. [6], respectively, it is seen that the interphase morphology observed in fiber-reinforced hybrid composites are similar to those for the neat polymers not only at all temperatures. This suggests that the interdiffusion mechanisms (swelling of the TP and diffusion of TS into TP) are similar. Nevertheless, since fibers prevent the thermoplastic from swelling freely and also act as an obstacle for TS monomers to diffuse, the fiber-reinforced hybrid composites exhibit a significantly lower interphase thickness at all temperatures, almost half of that exhibited by the neat polymers. The effect of fibers on the interphase thickness can also be seen in the non-constant interphase thickness of the hybrid composites. Due to the heterogeneous fiber distribution in the fabric, local fiber volume fraction of the polyester/glass layer changes from one location to another. This leads to a varying interphase thickness, where the regions with low fiber volume fraction exhibit a higher interphase thickness and vice versa. For instance, in Figure 4c, a clear gradient in interphase thickness can be seen. To the right of the cross-section shown in Figure 4c, there are stitching fibers (not seen in the figure) that decrease the local fiber volume fraction and eventually lead to a larger interphase thickness compared to the surrounding areas.

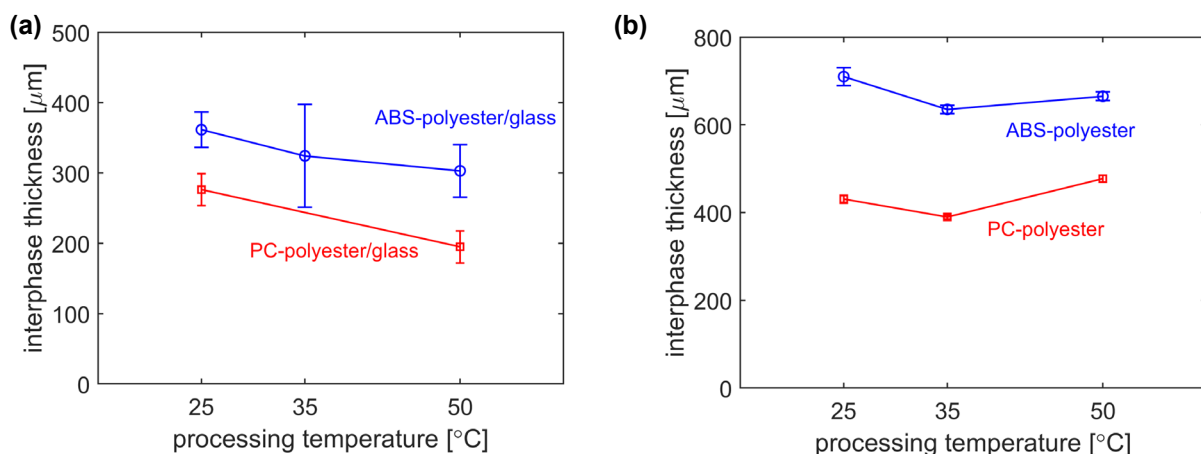


Figure 5: Interphase thickness of co-bonded (a) hybrid fiber-reinforced composites, (b) neat polymers. Data in (b) is reproduced from [6,7].

**Bond strength.** Force ( $F_d - F_w$ ) - displacement graphs of specimens tested via CDP test are presented in Figure 6. Graphs exhibit a zero-force region, which corresponds to the pre-crack, and a plateau region corresponding to crack propagation. Remarkable in the figure is the oscillatory force response about the plateau value, where the amplitude of the oscillations and the force level at the plateau depend on the type of the material and processing temperature. For instance, ABS-

polyester/glass exhibits more oscillations compared to PC and the amplitude of oscillations for this material tends to decrease with increasing processing temperatures. This trend can be explained by analyzing the photos of the fracture surfaces of the specimens and the optical micrographs of the crack path shown in Figure 7 and Figure 8, respectively.

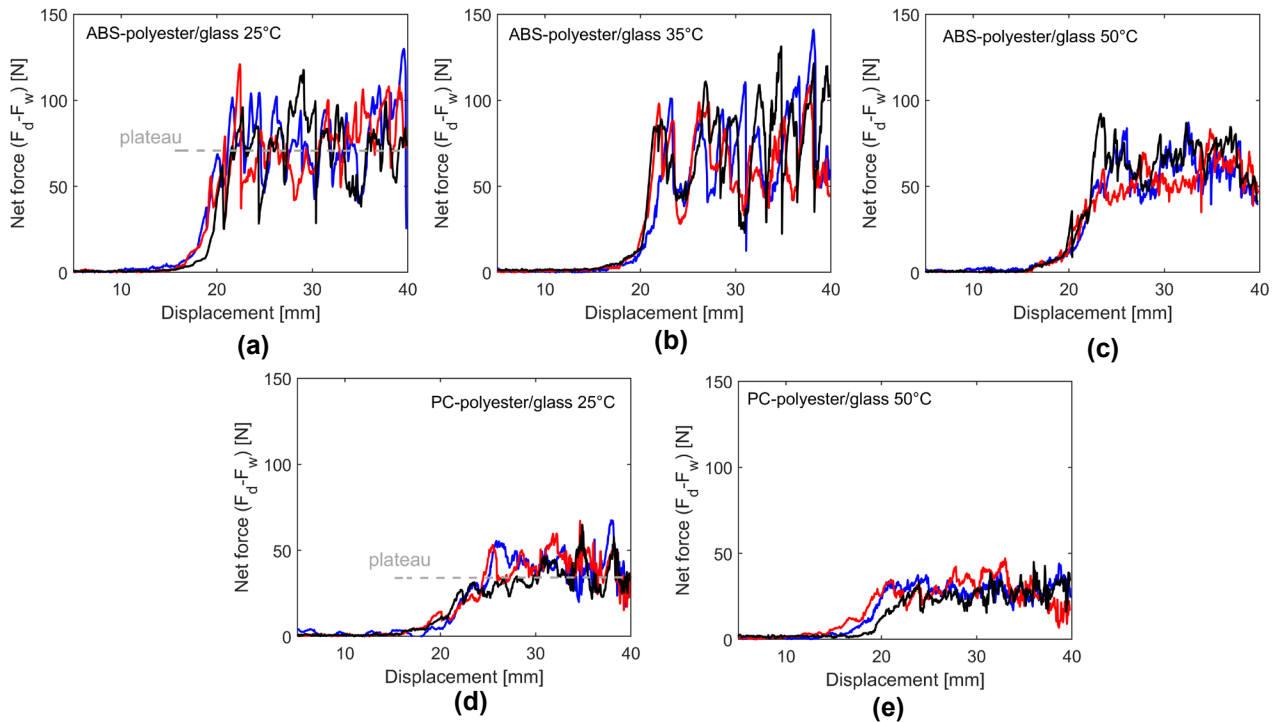


Figure 6. CDP force-displacement graphs of ABS-polyester/glass cured at (a) 25°C, (b) 35°C, (c) 50°C, and PC-polyester/glass cured at (d) 25°C, (e) 50°C. Data from three specimens are presented per test condition.

Figure 7 shows that there are white-colored remainders on polyester/glass skins of ABS-polyester/glass separated by the transverse fiber bundles. At room temperature, a high amount of remainders can be spotted on the polyester/glass skin. By analyzing the optical micrographs (Figure 8a and b) of the crack path for ABS-polyester/glass, it can be seen that the white-colored remainders are the remainders of the interphase. This shows that cohesive interphase failure plays a big role in the peel resistance of this specimen. Transverse fiber bundles are located on the ABS-side after fracture, indicating that extensive intra-ply failure takes place in addition to the aforementioned cohesive failure. Such intra-ply failure is thought to be responsible for the observed highly oscillatory force-displacement response [9]. With increasing temperature, the amount of remainders remarkably decreases, hence less interphase failure takes place. In addition, the number of transverse fiber bundles that remain on the ABS-side also decreases; hence, less intra-ply failure is observed at high temperatures. As seen in Figure 8c, at high cure temperatures, interface failure between the interphase and polyester/glass becomes prominent. A decrease in the extent of interphase and intra-ply failure at high temperatures correlate well with the lower force levels and amplitude of oscillation shown in Figure 6.

PC-polyester/glass cured at room temperature shows a failure behavior significantly different from that of ABS-polyester/glass: only a single fiber bundle can be detected on PC (Figure 7), which means that intra-ply failure does not play a significant role in peel resistance. This is also supported by the micrograph shown in Figure 8d which shows that the failure takes place at the interface of the interphase and polyester/glass. Interfacial failure also agrees with the smooth crack surface shown in Figure 7. At higher cure temperatures, failure is again mostly interfacial (Figure 7 and Figure 8e). Nevertheless, as shown in Figure 7, occasional intra-ply failure is not observed at 50°, unlike at 25°C. This means that the strength of the interface between the interphase and polyester/glass decreases at

high cure temperatures, promoting interface failure. More interfacial failure at 50°C correlates with lower force levels and oscillation amplitudes (Figure 6d and Figure 6e).

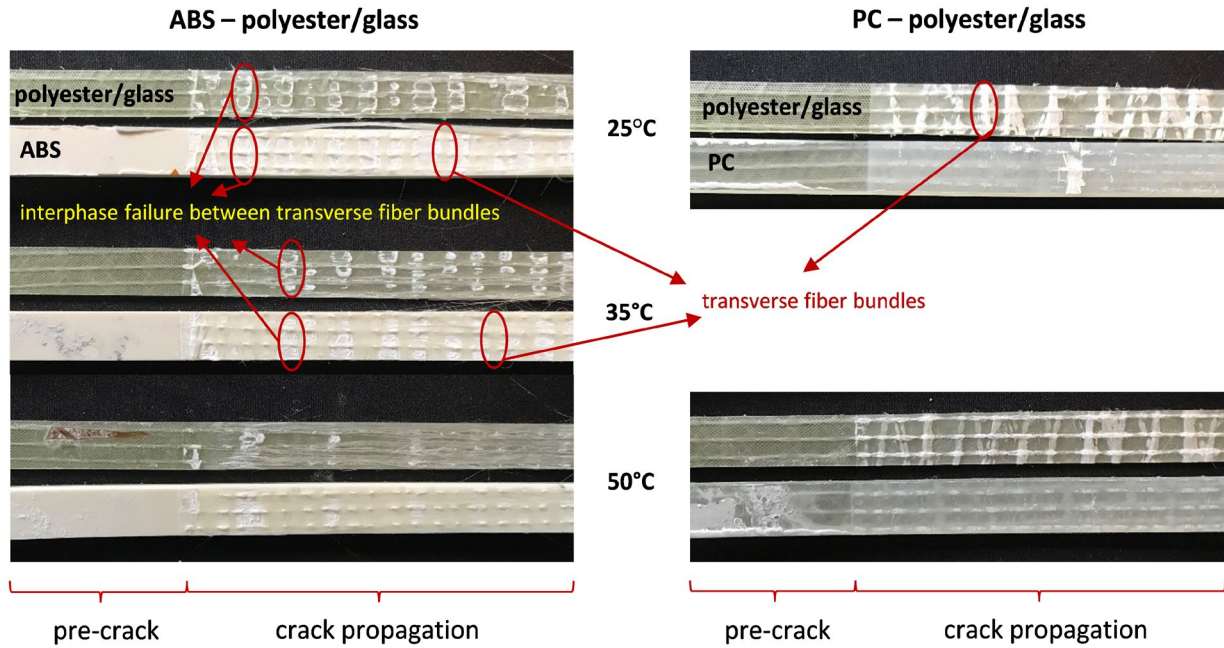


Figure 7. Fracture surface of (left) ABS-polyester/glass and (right) PC-polyester/glass cured at different temperatures.

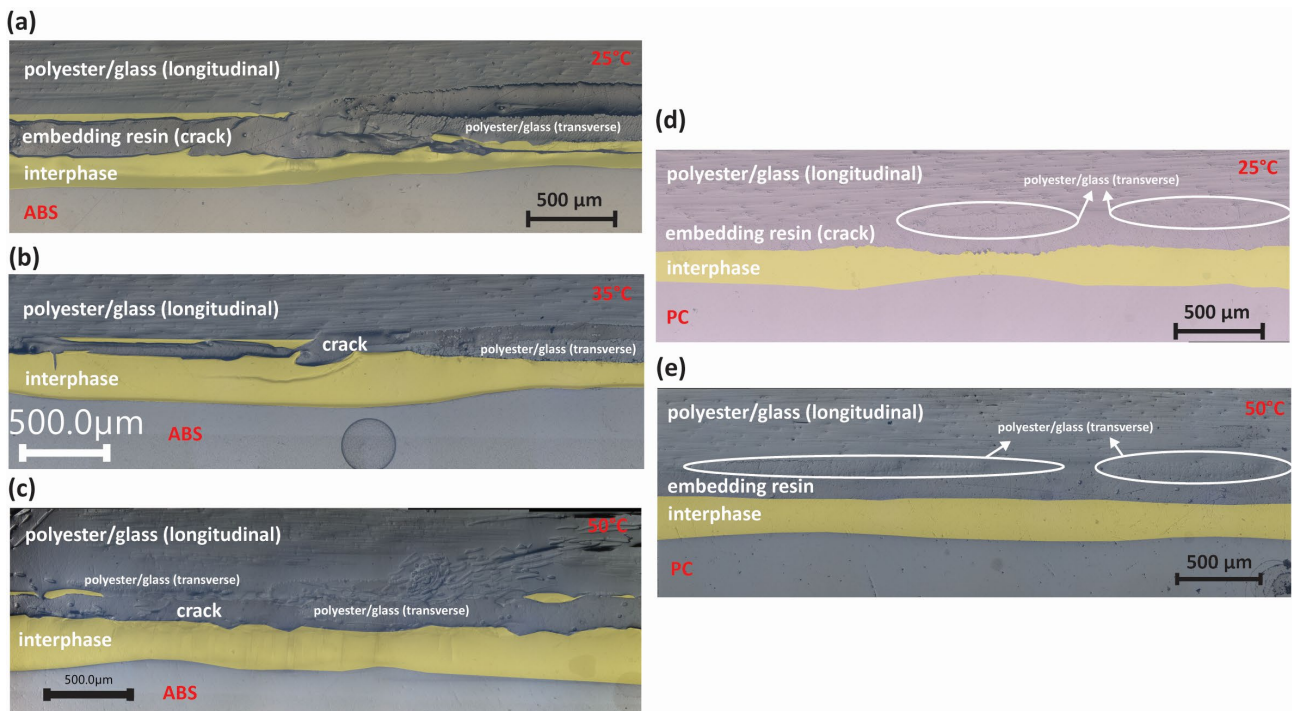


Figure 8. Optical micrographs of crack paths of ABS-polyester/glass cured at (a) 25°C, (b) 35°C, (c) 50°C, and PC-polyester/glass cured at (d) 25°C, (e) 50°C.

Next, peel energies of the two hybrid composites are presented in Figure 9 for different processing temperatures and their relation to interphase morphology is discussed. As implied by the force levels in Figure 6, peel energy decreases with increasing processing temperature. A remarkable finding is that the trend of peel energy vs. processing temperature (Figure 9) agrees with that of the interphase thickness of the composites vs. processing temperature (Figure 5a). Hence, a thicker interphase might imply a higher bond strength. A stronger bond promotes failure at the interphase and occasionally in

the intra-ply regions. However, when the bond is weak, failure tends to take place at or close to the interface of the interphase and polyester/glass, which is thought to be the weakest location across the interphase for both hybrid composites based on the fractography results.

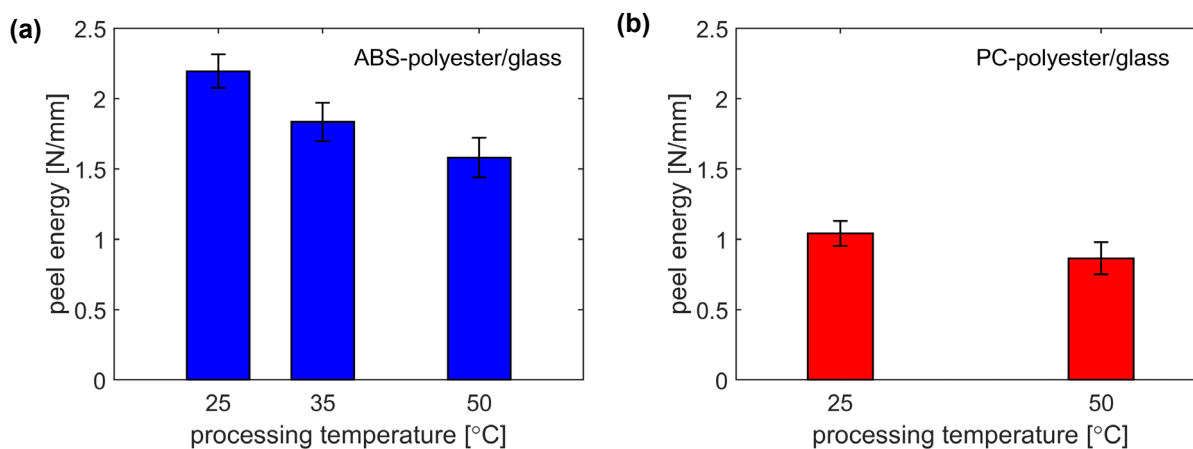


Figure 9. Peel energy vs. processing temperature for (a) ABS-polyester/glass and (b) PC-polyester/glass. Bar plots show the mean values and the error bars represent  $\pm 1$  standard deviation.

## Conclusions

In this work, the effect of cure temperature on the interphase morphology and the bond strength of ABS-polyester/glass and PC-polyester/glass hybrid composites was studied. Although the interphase morphology of the hybrid composites was found to be similar to that of the co-bonded neat polymers, the trend of interphase thickness vs. temperature was different. An increase in cure temperature led to a decrease in the interphase thickness for both hybrid composites. ABS-polyester/glass composites exhibited interphase and intra-ply failure, but the extent of those significantly decreased at high temperatures. At high cure temperatures, interfacial failure was prominent between the interphase and polyester/glass. Unlike ABS-polyester/glass, PC-polyester/glass exhibited interfacial failure (between the interphase and polyester/glass) both for curing at room temperature and 50°C. For both hybrid composites, an increase in temperature led to a decrease in the bond strength (peel energy). This correlated well with the decrease in the interphase thickness with increasing cure temperatures. At high temperatures, the interface (or the part of the interphase close to polyester/glass) between the interphase and polyester/glass was seen to be weaker, which may be linked with a decrease of interphase thickness signifying a lower level of interdiffusion.

The authors propose as future work the mechanical characterization of the interphase using nanoindentation and conducting further fractographic analysis with scanning electron microscopy. Furthermore, studying residual stresses is considered to be also important to understand the underlying factors in the fracture behavior of hybrid composites.

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