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Improved Interface Strength for Overprinting with CF-LMPAEK

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Abstract

Continuous fibre-reinforced PAEK laminates are attractive for aerospace structures, yet the integration of out-of-plane features requires costly secondary joining. In this study, the feasibility of in-situ overprinting of TC1225 prepreg laminates with short-fibre-reinforced LM-PAEK (Victrex 231GRA) using a robotic fused granulate fabrication (FGF) process is investigated. Two sources of heat are employed: a heated printbed and a LED-based interlayer heater (DEMEX), both individually and in combination. Substrates are also modified with a neat LM-PAEK film. T-pull-off tests are used to quantify the interfacial strength and fracture mode analysis distinguishes adhesive, mixed, and cohesive failures. Results show that interfacial strength increases dramatically with the substrate surface temperature. The highest values (~40 MPa) are obtained only when the printbed and DEMEX heaters are used in combination, achieving substrate surface temperatures of 300 °C. Failure modes shift from adhesive to predominantly cohesive at these temperatures, indicating effective fusion bonding. These findings demonstrate that a dual-heat source strategy is a reliable means of achieving robust overprinting bonds in semi-crystalline thermoplastic composites and paves the way for fully integrated hybrid laminate structures.

Keywords: *FGF; Overprinting; LM-PAEK; Additive Manufacturing*

1 Introduction

Continuous fibre-reinforced thermoplastic composites based on polyaryletherketone (PAEK) matrices are of great interest for structural aerospace applications, combining high specific stiffness, chemical resistance, and thermoplastic reprocessability [1]. Automated Fibre Placement (AFP) can be used to produce high-performance laminates from CF-PAEK efficiently, but is inherently limited in geometric complexity. Reinforcing structures like ribs, brackets, and other out-of-plane features typically require secondary joining operations, adding cost, weight, and potential failure sites. Overprinting, the direct deposition of extruded thermoplastic material onto a pre-consolidated laminate via material extrusion additive manufacturing, offers a route to in-situ hybrid structures that avoids these secondary steps.

Bond formation during overprinting is governed by molecular diffusion across the interface [2,3]. For semi-crystalline systems, the onset of crystallisation effectively terminates diffusion, creating a narrow bonding window that demands careful thermal control. Consul et al. demonstrated that fast-crystallising injection-moulding grades fail to develop meaningful interlayer strength in open environments due to crystallisation preceding the next layer deposition, while slow-crystallising PAEK grades achieve a dramatic improvement in Z-direction

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strength by extending the available diffusion window [4]. Critically, the authors concluded that where crystallisation cannot be delayed sufficiently through material selection alone, an external energy source is required to sustain the bonding window. This is particularly the case during overprinting of consolidated and thus fully crystallized laminates.

Overprinting with semi-crystalline high-temperature polymers is a particularly difficult task with few studies available in the literature [5,6,7,8]. In general, filament printing appears to be the most prominent technology, allowing for heated chambers in addition to energy input through a heated printbed and the extrudate itself. This, however, is not possible with robotic Fused Granulate Fabrication (FGF). To compensate this and further improve the bond quality, the extruder can be fitted with external heating systems like hot air [9], Laser [8], or most recently the LED-based DEMEX system from LEAM Technologies GmbH (Germany), promising both an extension of the process window and increased performance [10,11,12].

It is evident from the available literature above, that the contact temperature is one of the major drivers for fusion bonding. Tailoring the substrate surface by adding excess neat polymer may be beneficial as well. Carbon fibres near the bonding surface both decrease the available polymer-polymer bonding area and conduct heat away from the deposited extrudate. Thus, a neat polymer layer maximises the available bonding area [7] and reduces the cooling rate, acting as an insulation between extrudate and substrate [13].

This work presents a preliminary investigation of the effects of substrate surface type and temperature on the in-situ bond strength of single-walled stiffening structures applied with an FGF process and LM-PAEK matrix systems.

2 Materials and Methods

Substrates were manufactured from TC1225 prepreg (Toray Advanced Composites, Netherlands) stacked in a $[0, 90]_{2s}$ layup and vacuum consolidated on a heated table. Some substrates were modified by adding a neat LM-PAEK film (Victrex, UK) on both surfaces, thus avoiding warpage through asymmetry. The consolidated thickness was approximately 1.1 mm for prepreg only, and 1.2 mm with additional neat matrix. Substrate surfaces were wiped with isopropanol prior to overprinting to remove residuals from the UPILEX release film. Surface roughness was determined with a Keyence VR-5000 3D-Profilometer (Keyence, Japan) using the 12x magnification and wavelength filters $\lambda_s = 80\ \mu\text{m}$ and $\lambda_c = 2.5\ \text{mm}$.

The FGF process was performed with a DXR25 single screw extruder with a 2 mm round nozzle with a blunt tip (Weber, Germany) on a 6-axis industrial robot. The part was sliced with a layer height and width of 0.5 and 3 mm, respectively. The geometry features a 170 mm straight line transitioning into a 25 mm radius turn, resulting in a layer length of 497 mm (Figure 1). Kapton tape was used to mount the substrate on the printbed. This allows for slight deformation from crystallization and CTE mismatch and thus decreases the residual stresses in the bonding zone.

The material used was short-fibre reinforced LM-PAEK pellets of type 231GRA (Victrex, UK), which were dried at 130°C for at least 12h prior to extrusion. For all prints, the extruder temperatures were held constant, resulting in a material extrusion temperature of 361°C. This was measured by inserting a 250 µm thermocouple into the nozzle during extrusion. The substrate surface temperatures were measured with 80 µm thermocouples placed on the surface[1]. Additional data was exported from the DEMEX IR-camera log, using the median

[1] Extruder and substrate temperatures are considered static and dynamic in nature, respectively, hence the differing thermocouple gauges used to capture information with the appropriate temporal resolution.

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temperature of the 10 highest value pixels in the Stage 1 zone, just ahead of the nozzle (with respect to its printing direction/motion), with an emission coefficient of 1.0. The setpoint temperature for the DEMEX heating was fixed to 300 °C where applicable. Table 1 lists the printing parameters relevant to the interfacial temperatures and the average power level of the DEMEX LED in the straight section during the first layer.



Figure 1: Printed structure on the printbed (left) and T-shape coupons (right).

Tensile pull-off tests were used to assess the mechanical strength of the overprinting bond. For each configuration, T-shaped coupons of dimensions 45 x 20 x 50 mm (L W H) were cut from the printed structure using a water-cooled circular diamond blade. Machining of the printed walls was disregarded in favour of fast testing, thus accepting possible eccentric loads due to clamping of rough surfaces. Mechanical testing was performed with a tensile testing machine and a T-pull-off rig build from two horizonal steel plates of 10 mm thickness with a gap of approximately 6 mm. The traverse speed was set to 1 mm/minute, with forces recorded on a 5 kN load cell. The ultimate stresses at failure were calculated using the cross-sectional area of the printed wall (i.e. bead width). This was chosen from the perspective of a design task, where stresses should be related to a designable cross-section. For a single-walled structure in an FGF process with uniform extrusion, the bead width is the only major parameter.

Table 1. Printing conditions related to overprinting temperature

Heating	Print speed first layer (mm/s)	Substrate Surface Temperature (°C)	DEMEX Setpoint (°C)	DEMEX power on first layer (%)
Printbed only	60	140	n.a.	n.a.
	60	178		
	60	190		
DEMEX only	15	22	300	100^{T} / 100^{L}
	30	22	300	100^{T} / 100^{L}
	60	22	300	100^{T} / 100^{L}
Printbed and DEMEX	15	220	300	30.4^{T} / 31.2^{L}
	60	193^{T} / 220^{L}	300	90.7^{T} / 78.8^{L}

Values for TC1225 surface indicated with ^{T} and for LM-PAEK surface with ^{L}

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3 Results and Discussion

3.1 Substate Lamiantes

The substrate surface analysis is shown in Figure 2. The surface roughness was determined as the average of 41 parallel lines, as indicated in the centre image. The matrix-rich surface (LM-PAEK surface) exhibits an increased surface roughness of 11.34 µm compared to 6.17 µm for the prepreg surface. While at first counterinterintuitive, microscopy of the samples indicated fibre migration from the laminate surface into the neat matrix film, with fibres occasionally emerging on the surface. The prepreg surface, by comparison, exhibits some regions with matrix accumulation resulting from the vacuum consolidation, though typically not more than 10 µm (in the through-thickness direction).

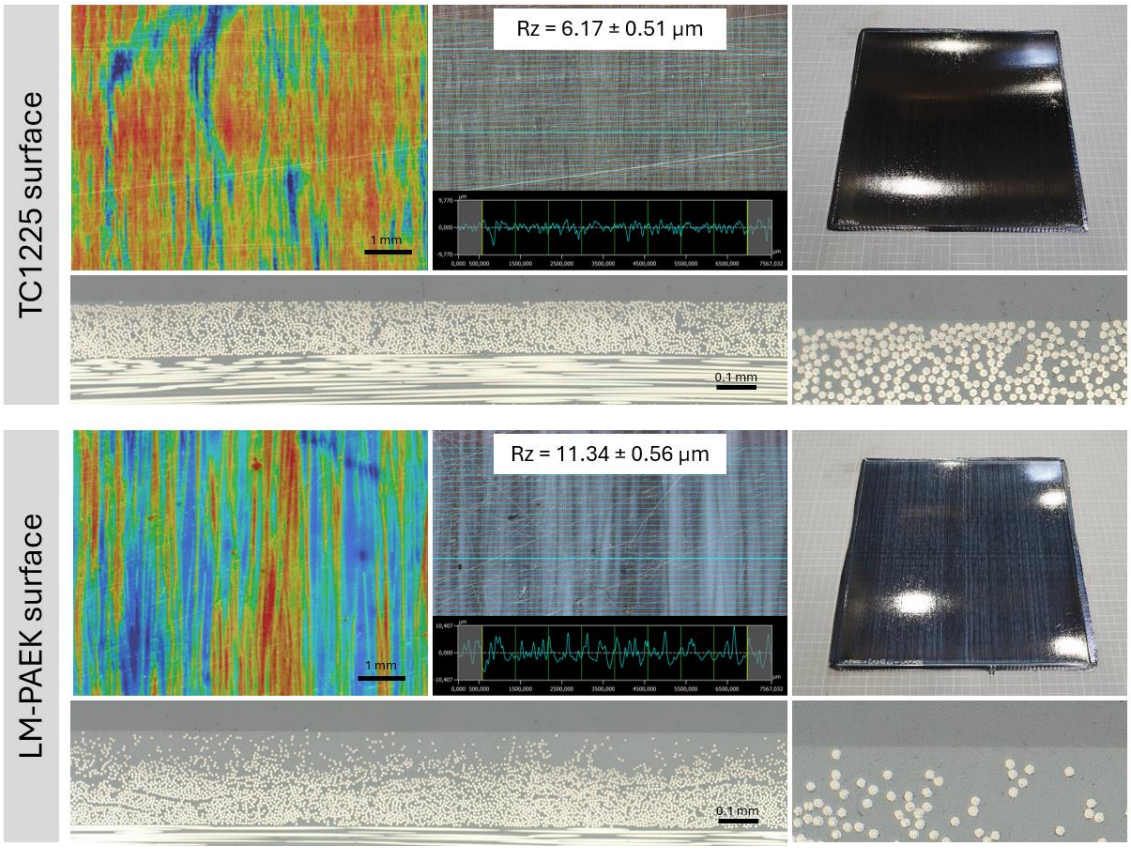


Figure 2: Substrate surface inspection with a colormap of the height profile (left), optical profilometer image and surface roughness (centre), macro image of the substrate laminates (right) and micrographs (bottom) for both surface conditions, respectively

3.2 Tensile Pull-Off Test

Figure 3 contains box plots of the ultimate stresses at failure for all heating conditions, with blue indicating TC1225, and orange indicating matrix-rich LM-PAEK surfaces. For the case of DEMEX only, the print on TC1225 surfaces with 30 and 60 mm/s failed during sample cutting and is thus excluded. All cases exhibit significant scatter, which is common for additive manufacturing but may be particularly prominent due to some eccentric load associated with clamping of raw print surfaces. When only printbed heating was employed, the interface strength increased strongly with the substrate temperature. This was most clearly visible for the matrix-rich surface samples. The prepreg surface can be assumed to behave similarly based on available data, though the reduced number of datapoints and a less prominent gradient make the trend more difficult to

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observe. The matrix-rich surface samples also interacted strongly with DEMEX heating. As a reduced print-speed correlates with a higher temperature, the trends observed using the independent heat sources are consistent with one another. The combination of both heating methods leads to a strong increase in performance for both surface types. Interestingly, the dependency using this heating configuration was more visible for the prepreg surface than the matrix-rich surface. The decrease in strength for TC1225 and 60 mm/s was likely influenced by the lower substrate surface temperature reported in Table 1, which was caused by the slow response of the heated printbed and this sample being the first print, before full equilibration.

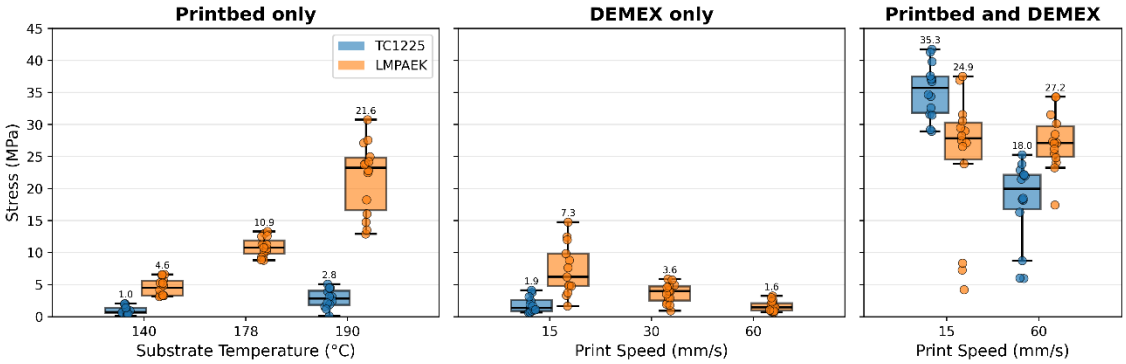


Figure 3: Ultimate stress at failure for the T-pull-off test

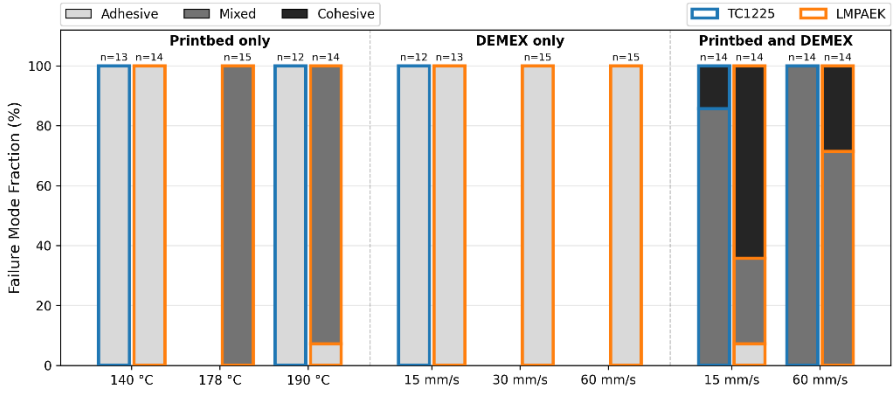


Figure 4: Distribution of failure modes

Figure 4 details the distribution of failure modes observed within the tests. Three distinct failure modes were observed in the mechanical test campaign: fully adhesive, mixed (some printed material remaining on the substrate), and fully cohesive mode (fracture exclusively in the printed structure). Higher values of tensile strength correlate with a larger fraction of mixed or cohesive failure modes. Fully cohesive failure modes were only observed with combined printbed and DEMEX heating. Mixed failure modes often exhibited cohesive regions with a diameter equal to the nozzle diameter. This suggests a varying degree of bonding across the bead width, correlating with the hydrodynamic pressure under the open nozzle. An overview of representative fracture surfaces is given in Figure 5.

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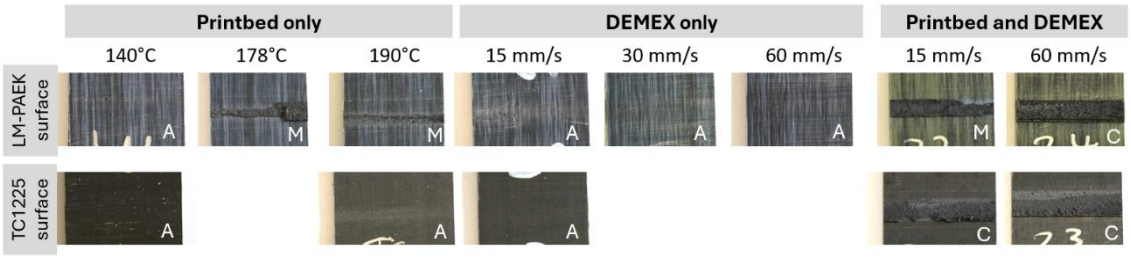


Figure 5: Representative fracture surfaces with classification as Adhesive, Mixed or Cohesive failure

3.3 Influence of Substrate Temperatures

The temperature of the substrate surface increases throughout the heat affected zone, up to the edge of the nozzle. The section of the DEMEX control zone just in front of the nozzle in the direction of travel ("Stage 1" zone) is used for temperature control. This reading is considered the substrate temperature at the moment of overprinting and is displayed on the left side of Figure 6. For the case of DEMEX only, the TC1225 substrates result in consistently higher temperatures. This is likely due to the variance of the absorption and emission coefficients, leading to different real and apparent temperatures, respectively. For lower print speeds, the Stage 1 temperature increased due to longer exposure times. The setpoint of 300 °C was not reached unless printbed heating was employed. This points to insufficient heating power of the LEDs superimposed with thermal diffusion within the substrate. This is in line with the LED power readings given in Table 1. The right diagram in Figure 6 provides an overlay of temperature readings and the average stress at failure for each condition. One can observe a positive trend, which is in line with the expectation based on polymer bonding theory. The data suggests that the overall low interface strength is a direct result of a lack of temperature of the substrate surface. The fact that samples with only printbed heating performed better than those with only DEMEX heating, despite similar temperatures, may be due to the full volume heating in the former case, compared to more surficial heating in the latter. This would lead to fast cooling rates and thus less effective time for diffusion.

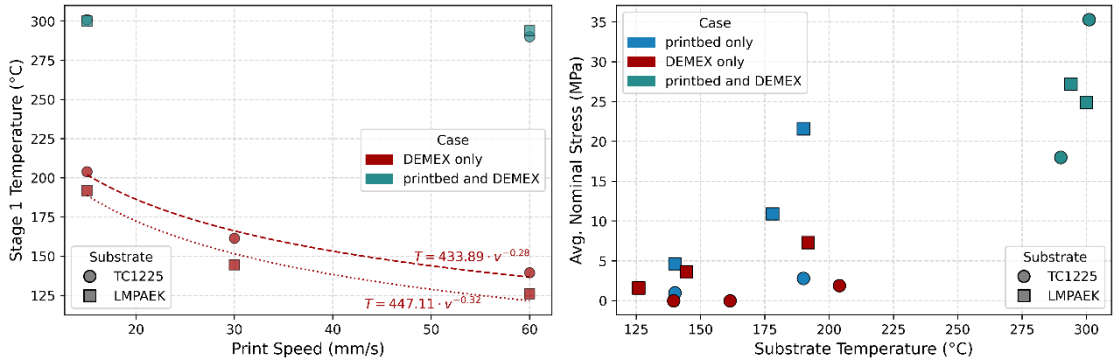


Figure 6: Stage 1 temperature as a function of print speed (left) and increase of overprinting bonding strength with substrate temperature by means of printbed and DEMEX heating (right)

3.4 Part Deformation

The combination of CTE mismatch between the short fibre-reinforced printed structure and continuous fibre laminate, and the one-sided crystallization shrinkage in the printed structure only, can be expected to result in residual stresses within overprinted structures. The method of loosely mounting substrate laminates to the heated table employed here allows for some reduction in stresses by deformation. Figure 7 shows a profilometer scan of the substrate backside after overprinting. The laminate exhibited a double curvature about the axes of

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symmetry. The diagram on the right side quantifies the horizontal centreline profiles of deflection for combined DEMEX and printbed heating.

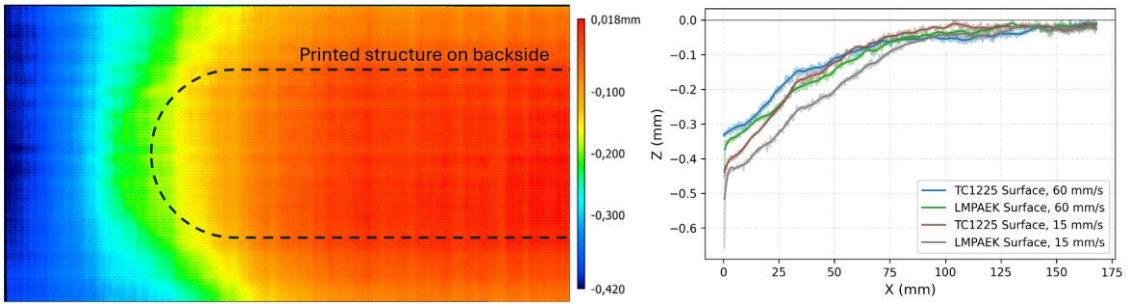


Figure 7: Profilometer scan of the left half of the substrate backside after overprinting (left) and profiles of longitudinal deflection in the horizontal centreline

4 Conclusion and Outlook

This paper has demonstrated the feasibility of in-situ overprinting of laminates with an FGF process and LM-PAEK matrix systems. The substrate surface temperature is one of the main factors influencing the interface strength. This temperature can be conveniently manipulated by means of a heated printbed and an external extruder-mounted heat sources, specifically the temperature-controlled LED heating system DEMEX from LEAM Technologies GmbH. The results indicate a strong trend of increasing interface strength with increasing surface temperature. The observations suggest that the necessary temperatures cannot be reached by any of the two heat sources alone, but only in combination. It was shown that good printing conditions can lead to predominantly cohesive failure. Further steps in the process development for overprinting should focus on the definition of ideal temperature conditions and the quantification of the process window for a given combination of heat sources and materials.

Acknowledgements

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