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# TITLE: Interface Formation and Bonding Mechanisms in Overprinting of Thermoplastic Composites: Process Parameters, Surface Treatment, and Diffusion Kinetics

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**ABSTRACT:**

Overprinting is a rapidly emerging hybrid manufacturing process that combines the high specific strength of thermoplastic composites with the design flexibility of Additive Manufacturing (AM), by extruding polymer features directly onto thermoformed composite substrates. However, the structural integrity of these hybrid parts relies entirely on the quality of the interfacial bond formed between the extruded filament and the consolidated composite substrate. This study investigates the surface texture induced by the flow processes and bonding mechanisms governing this interface. We present a comprehensive analysis of overprinting high temperature polyamide (PAHT) and polyaryletherketones (PAEK), examining the influence of extrudate flow driven by nozzle settings, surface energy modification via plasma treatment, and the competition between molecular diffusion and crystallisation kinetics.

Two material systems were investigated: a high-temperature polyamide (PAHT) printed onto CF/PA6 substrates, and a semi-crystalline PAEK (Victrex AM™ 200) printed onto CF/PAEK composites (Victrex AE™ 250). The bonding quality was quantified using a rib-on-plate tensile test method designed to isolate interfacial failure.

The study explored three distinct optimisation strategies: 1) Process Parameters: Varying nozzle offset heights *h* and layer thicknesses *t* to alter the pressure distribution and contact area during the deposition flow [1]. 2) Surface Modification**:** Application of atmospheric plasma treatment to modify the surface free energy and wettability of the composite substrate prior to printing. 3) Thermal History and Kinetics: Development of a modified non-isothermal healing model [2] to predict bond strength based on the thermal history at the interface, specifically accounting for the competition between molecular reptation and crystallisation.

For the PAHT/CF/PA6 system, the flow of the extrudate and its ability to wet the composite surface were heavily influenced by the nozzle offset height *h*. Reducing the nozzle offset from 0.2 mm to 0 mm or -0.2 mm significantly improved the bonding force. When *h*=0 mm or lower, the nozzle effectively promotes squeeze flow of the molten filament into the weave composite texture, increasing the contact area (i.e. line width and roughness) and applying pressure that facilitates wetting. Fractographic analysis revealed that specimens with high bond strength exhibited mixed cohesive/adhesive failure modes, characterised by residual resin and fibre transfer on both fracture surfaces, whereas weak bonds displayed clean adhesive failure.

To further enhance the flow and wetting of the polymer melt without relying solely on mechanical pressure, plasma treatment was applied to the CF/PA6 substrates. The treatment increased the surface energy, improving the wettability of the composite. This treatment resulted in a significant increase in bond strength. This suggests that optimising surface energy is critical for facilitating the necessary wetting flow before the polymer melt solidifies.

For the high-performance semi-crystalline PAEK system, the study focused on the molecular flow across the interface. A modified non-isothermal healing model was developed that accurately describe the competition between reptation and crystallisation. It was identified that crystallisation inhibits diffusion: once the polymer crystallises, molecular mobility ceases. Consequently, the interfacial bond strength is determined by the effective diffusion time, which is the duration that the interface remains above the crystallisation onset temperature but below the melting point. Interestingly, the study found that overprinted PAEK joints achieved higher strengths compared to traditional overmoulded counterparts reported in literature [3]. This

is attributed to the lower pressure of the AM process, which, unlike high-pressure injection moulding, does not accelerate the crystallisation onset, thereby allowing a longer window for diffusion flow.

A positive linear correlation was established between the calculated final degree of healing and the true interfacial strength, validating the model as a predictive tool for diffusion-based bonding in semi-crystalline polymers [2].

This work demonstrates a comprehensive picture of interfacial flow and autohesion in overprinting of thermoplastic composites. Mechanically, ensuring intimate contact through optimised nozzle pressure (offset height) is essential for bonding. Additionally, increasing surface energy via plasma treatment significantly aids the wetting flow. Thermally, the bond formation is governed by the competition between molecular diffusion and crystallisation kinetics. By controlling these flow and thermal processes, overprinting can be optimised to produce robust, functionalized composite structures.

**References:**

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