Beyond Melting: Amorphous Bonding for Joining and Consolidation

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ABSTRACT

Crystallization may be the hidden constraint in thermoplastic composite manufacturing. It requires tightly controlled cooling, induces residual stresses through shrinkage, and introduces path-dependent behavior that complicates predictive modeling yet remains essential for structural performance. This work asks: can bonding be achieved without relying on melt-driven crystallization?

To address this, thin (5–20 μm) polyetherimide (PEI) interlayers are pre-healed to slow-cooled carbon fiber reinforced polyaryletherketone (PAEK) in two contexts. The first, Thermabond, is sub-melt joining of low melt-PAEK laminates. Results show that bond quality is governed primarily by processing (i.e., adequate healing and film handling) rather than modest changes in interlayer thickness.

This concept is then extended to laminate-scale manufacturing through an architecture known as OATMEAL (Out-of-autoclave Amorphous/semicrystalline Thermoplastic Material for Energy-efficient Aerospace-grade Laminates). PEI is bonded to carbon fiber reinforced polyetheretherketone (PEEK) prepregs (i.e., tapes) and excess PEI is then laser ablated from the surface. Crystallinity is developed off-line during prepreg fabrication, while subsequent consolidation occurs below PEEK's melt temperature to preserve it. Cross-ply warpage experiments show that, contrary to intuition, amorphous interfaces, i.e., PEI, reduce global curvature by lowering the effective stress lock-in temperature and eliminating crystallization shrinkage from the lamina response. Correspondingly, laminate behavior is accurately predicted using the classical lamination theory (CLT) with a single effective stress-free temperature, whereas conventional carbon fiber PEEK laminate requires accounting for crystallization-driven effects.

By decoupling interfacial healing from crystallization, OATMEAL enables sub-melt consolidation, reduces energy consumption by up to 75%, and increases manufacturing throughput by fivefold. These results of this study demonstrate that amorphous bonding is not only a joining strategy, but a pathway to more predictable and scalable thermoplastic composite manufacturing.

Keywords: thermoplastic composites, polymer healing, fusion bonding, OATMEAL
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INTRODUCTION TO AMORPHOUS BONDING

The high energy consumption and long cycle times associated with thermoplastic composite manufacturing remain a key barrier to achieving the production rates required for next-generation aerospace structures [1]. Semi-crystalline systems such as CF/PEEK, CF/ polyetherketoneketone (PEKK), and CF/low-melt polyaryletherketone (LM-PAEK) offer excellent structural performance, weldability, and chemical resistance, but their processing remains constrained by tightly coupled thermal and crystallization phenomena [2]. Across automated fiber placement (AFP), welding, and out-of-autoclave consolidation, the challenge is not only achieving intimate contact, healing, and consolidation within a limited thermal cycle, but doing so in a way that is predictable and robust.

Thermoplastic fusion bonding is typically described as a sequence of intimate contact, polymer healing (also known as interdiffusion or autohesion), and solidification [3]. In amorphous polymers, healing can proceed above the glass transition temperature, T_g, providing a relatively broad processing window [4]. In semi-crystalline systems, crystallization is often cited as limiting healing [5]; however, in high-rate processes, the more significant issue is that there is insufficient time for crystallization and relaxation to fully develop. As a result, morphology and residual stress become strongly dependent on the exact thermal history. This creates a coupled, path-dependent process in which healing, crystallization, and shrinkage must all be considered simultaneously complicating both process design and modeling [6].

From an engineering standpoint, this coupling limits predictability. Accurate modeling requires resolving crystallization kinetics, melt memory, and their interaction with thermal gradients and pressure, physics that are difficult to measure and even more difficult to control in manufacturing environments. A more desirable approach is to decouple these effects so that bonding can be governed by a simpler and more controllable mechanism.

Amorphous interlayers provide one such pathway. By shifting interfacial healing to an amorphous polymer, bonding can occur above T_g without requiring the surrounding semi-crystalline material to fully melt [7], [8], [9], [10], [11]. This separates interfacial strength development from bulk crystallization, reducing the problem to polymer interdiffusion at the interface and significantly reducing sensitivity to thermal history. The Thermabond® process provided a firm foundation when Cogswell and team introduced thick polyetherimide (PEI) interlayers to enable sub-melt joining of PEEK laminates via welding, demonstrating that amorphous bonding can achieve strengths comparable to melt-based joining and adhesive alternatives while preserving crystallinity [7], [8]. However, these systems relied on resin-rich interlayers on the order of ~200 μm, which reduce fiber volume fraction and degrade structural efficiency. While acceptable for laminate-to-laminate joining, this becomes a limiting factor when extending the concept to prepreg-level architectures, where the cumulative amount of amorphous material becomes critical [12].

Out-of-autoclave amorphous/semi-crystalline thermoplastic materials for energy-efficient aerospace-grade laminates is named OATMEAL for its Goldilocks zone processing.

OATMEAL builds on this foundation by reducing the amorphous interlayer to the micrometer scale through laser ablation [9]. The architecture also simplifies the governing physics: consolidation occurs within a "Goldilocks" window above the PEI T_g and below the PEEK melting temperature, where interfacial healing dominates and crystallization is effectively removed from part manufacturing (see Figure 1).

This simplification has direct implications for engineering predictability. In the present work, cross-ply [$0_2/90_2$] OATMEAL laminates exhibit warpage that is accurately captured using classical laminate theory (CLT) with constant material properties and a single effective stress lock-in temperature. In contrast, equivalent CF/PEEK laminates cannot be predicted with the same approach, as crystallization-induced shrinkage and path-dependent behavior are not captured in standard CLT formulations. The OATMEAL architecture therefore enables both simpler processing and simpler modeling.

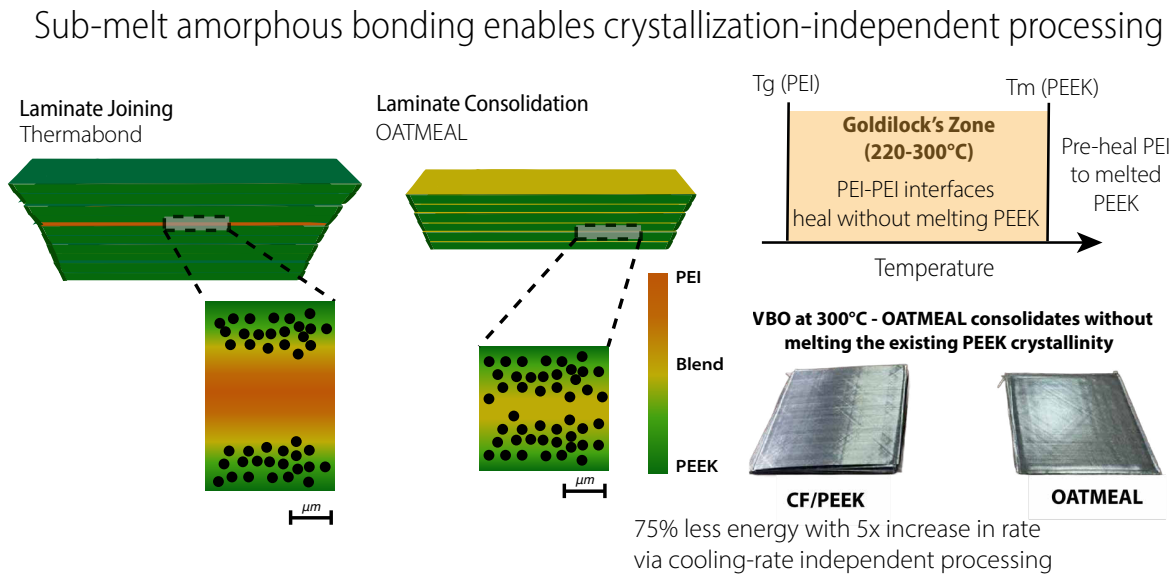

Figure 1. Thermabond® uses amorphous bonding to create a Goldilocks zone (above amorphous polymer Tg and below semi-crystalline polymer Tm) for laminate joining whereas OATMEAL applies it at the prepreg level to enhance part manufacturing. Adapted with permission from Advanced Materials, Kirchhoff et al., 2026, © Wiley [9].

To explore this behavior, the present study examines amorphous bonding across two manufacturing scales. Thermabond® is first treated as a controlled joining problem, isolating the mechanics and process sensitivity of a single amorphous bondline with varying PEI thickness. OATMEAL then extends this concept to full laminate consolidation. This raises the central question:

Can decoupling crystallinity from fusion bonding improve structural predictability, increase manufacturing throughput and maintain high-performance?

The results show that amorphous bonding is not merely a joining method, but a practical manufacturing strategy. By decoupling interfacial healing from crystallization, it enables more predictable processing, reduces sensitivity to thermal history, and allows standard engineering tools such as CLT to accurately capture laminate behavior. Finally, results from a fast vacuum-bag-only (VBO) process—*where parts are removed directly from the oven at 300°C without controlled cooling*—demonstrate that OATMEAL provides a pathway toward high-rate, energy-efficient thermoplastic composite manufacturing.

JOINING VIA THERMABOND

Thermabond® provided an ideal starting point for studying amorphous interfacial bonding, as it isolates the problem to a single interface using thin (5 μm) PEI films. Two pre-consolidated LM-PAEK laminates were joined via this PEI interlayer to address a practical manufacturing question prior to extending the concept to OATMEAL: at realistic, thin bondline thicknesses, is joint performance sensitive to the presence and thickness of the interlayer itself, or to whether the PEI is sufficiently processed to achieve effective healing?

While LM-PAEK is not the ideal semi-crystalline matrix (its lower melting temperature narrows the processing window and compresses the effective "Goldilocks zone" for sub-melt bonding) it provides a sufficiently representative platform for initial investigations into interfacial healing behavior. The semi-crystalline prepreg used to simulate Thermabond® was unidirectional carbon fiber–reinforced low-melt polyaryletherketone (LM-PAEK™, $T_m \approx 303$°C), supplied by SUPREM, with nominal dimensions of 6.35 × 0.19 mm and a fiber volume fraction of ~55%. Polyetherimide (PEI, Ultem 1000) was introduced as a thin amorphous interlayer using 5 μm-thick films (Piezopvdf). High-temperature vacuum bagging materials were sourced from Airtech, and steel tooling was prepared with Frekote mold release. A two-step vacuum bagging process was used to decouple processing from bondline thickness. In the first step, "half-plates" (96 × 160 mm) of unidirectional $[0]_{14}$ LM-PAEK were consolidated at 360°C for 30 minutes with PEI applied to the tool-side surface, followed by slow cooling in the oven to maximize crystallinity. PEI thickness was varied from 5–20 μm by stacking films, and the resulting plates were sectioned into 32 × 160 mm strips.

In the second step, pairs of half-plates were bonded to form $[0]_{14}$/PEI/$[0]_{14}$ laminates. Bonding was performed at 230°C, 280°C, and 360°C, followed by air cooling to simulate the rapid thermal cycles typical of high-rate processes (e.g., automated fiber placement and thermoforming). Since the PEI glass transition temperature is $T_g \approx 216$°C, all bonding conditions allow interfacial healing, while the LM-PAEK remains below its melting temperature for the two lower-temperature cases.

Processing dominates bond quality

The SBS results in Figure 2 are more informative of bond quality than of film thickness. Representative force-displacement curves highlight how premature failure of the PEI interlayer, which is common in the 230°C and 280°C samples, can be misleading if peak load is interpreted

naively (see Figure 2A). Once the PEI interlayer fails, the LM-PAEK half-plates can continue carrying load and deform plastically before final failure. Although ASTM D2344 recommends defining peak load using a 30% load drop, a 10% drop was used to better capture the initial failure associated with the PEI interlayer. For example, in the 230°C, 5 μm sample (Figure 2, left, blue curve), the SBS is 105.40 MPa using the 30% criterion but only 78.66 MPa using the 10% criterion. The larger value more closely reflects continued deformation and failure in the LM-PAEK half-plates, whereas the 10% criterion better isolates the healing-limited response of the PEI interlayer. In Figure 2B specimens that failed prematurely in the PEI are marked with stars. As processing temperature increases, the dominant failure mode shifts from premature PEI failure toward more complex plastic deformation and delamination in the LM-PAEK (see Figure 2C). Even at 280°C, however, some specimens still fail prematurely, particularly those with thicker PEI interlayers, suggesting that adding more PEI may become detrimental when healing remains incomplete.

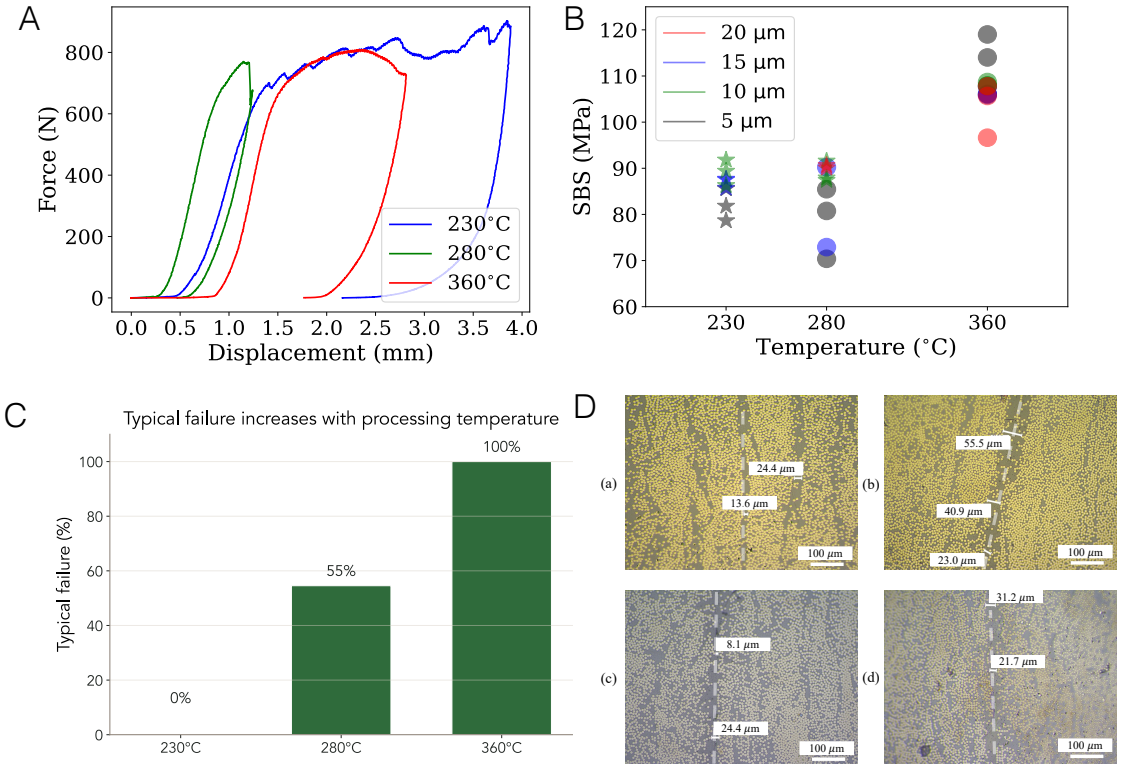

Figure 2. A: SBS force-displacement curves, B: SBS results with stars showing premature failure in the PEI., C: Failure mechanism trend as temperature increase, D: Cross-sectional Microscopy of (a) 360°C 5-μm, (b) 360°C 20-μm, (c) 230°C 5-μm, (d) 280°C 10-μm PEI.

Cross-sectional imaging was used to quantify the final PEI interlayer thickness and assess the extent of interdiffusion. Representative micrographs (Figure 2D) show that increasing the nominal PEI thickness on the half-plates generally leads to a thicker bondline in the consolidated laminate. During half-plate processing, the LM-PAEK is fully melted, allowing the PEI and PEEK to interdiffuse, with some fiber repopulation into the PEI-rich region. This effect becomes more pronounced in the full laminates processed at 360°C, where the LM-PAEK is remelted. In

these samples, fibers are observed throughout the amorphous bonding region, which likely contributes to the improved short-beam shear (SBS) strength.

Despite this, significant variability in interlayer thickness is observed. For example, in the 20 μm, 360°C condition, the final bondline thickness ranges from approximately 20 to 50 μm (Fig. 4D), indicating nonuniform PEI distribution. This variability is consistent with the strong temperature dependence of PEI healing kinetics reported by Barroeta Robles et al., who found that PEI–PEI welding times decrease by orders of magnitude with modest temperature increases, from 10^2 minutes at 280°C (63–321 min depending on interfacial conditions), to ~ 1 minute at 300°C, and to sub-second timescales by 320°C [11]. Such sensitivity highlights how small variations in thermal history can lead to large differences in interfacial healing, and therefore bond uniformity.

The key insight from this Thermabond® study is that while a thin amorphous interlayer can enable sub-melt joining, bond quality is governed primarily by thermal history rather than interlayer thickness. In the context of joining, PEI thickness is therefore a secondary parameter; however, at the prepreg level in OATMEAL, it becomes critical for maintaining fiber volume fraction and structural efficiency. This distinction clarifies the role of LM-PAEK as a model system: while it is effective for isolating single-interface bonding phenomena, its relatively low melting temperature restricts access to the higher-temperature regimes where PEI healing is most rapid. This motivates the transition to PEEK-based OATMEAL architectures, where bonding can reliably occur within an expanded "Goldilocks" zone near 300°C.

Key Lessons

- **Healing governs failure**
Incomplete interdiffusion means premature interlayer failure.
- **Thickness (and residual stress) is secondary**
Only matters once sufficient healing is achieved.
- **Material choice + handling are critical**
LM-PAEK limits access to rapid healing regimes (~300°C), while 5 μm PEI films are difficult to handle (wrinkling, nonuniformity), reducing quality.

LAMINATE CONSOLIDATION VIA OATMEAL

Based on these insights, the OATMEAL approach leverages the full miscibility of PEI and PEEK ($T_m \approx 343$°C) to form a coherent interphase between a semi-crystalline CF/PEEK substrate and an amorphous PEI layer. Hybrid OATMEAL prepreg is fabricated by sandwiching a CF/PEEK ply (Toray Cetex TC1200, ~125 μm thick) between 25 μm PEI films (CS Hyde), followed by consolidation at elevated temperature to ensure complete interdiffusion and bonding. This initial processing is intentionally conservative—using standard thermoplastic consolidation conditions—to establish a well-bonded baseline. Further details of the prepreg fabrication and material system are provided in our prior work [9].

A key design parameter in OATMEAL is the thickness of the amorphous PEI layer. The Thermabond® study showed that ultra-thin (5 μm) films are difficult to handle, tending to

wrinkle and introduce variability. For OATMEAL, thicker (25 µm) PEI films were used to ensure uniform coverage and manufacturability, with the understanding that excess amorphous content lowers fiber volume fraction and can diminish structural performance. To reconcile these competing requirements, laser ablation was employed as a scalable method to precisely tailor the interlayer thickness.

Laser ablation provides rapid, non-contact, and spatially controlled removal of the PEI layer, and has been previously demonstrated for composite surface modification at NASA Langley [13]. In this work, a picosecond UV laser is used to selectively remove PEI from the prepreg surface, enabling micron-scale control of interlayer thickness with minimal thermal impact to the underlying composite. Figure 3a illustrates the PEI–PEEK healing step at 380°C followed by laser ablation, while Figure 3c shows representative laser confocal measurements of the resulting surface topography. This "add-then-subtract" strategy (depositing excess PEI for robust handling and uniformity, followed by controlled ablation) enables the fabrication of prepreg that can be processed below the melt temperature while maintaining sufficient interfacial material to promote void-free consolidation. Ongoing internal R&D efforts at NASA Langley are focused on scaling this approach from lab-scale demonstrations to roll-to-roll manufacturing.

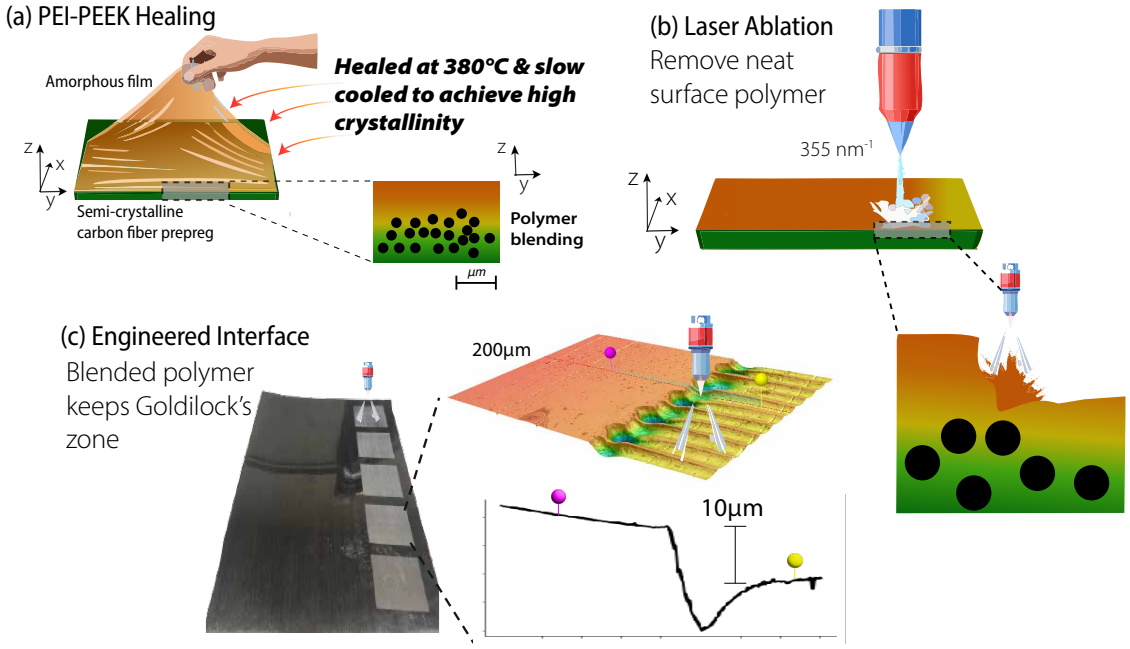

Figure 3. OATMEAL prepreg is made via (a) slow cooling after healing PEI film to CF/PEEK prepreg, (b) laser ablating surface patterns to remove excess PEI (c) while creating grooves for void removal. Adapted with permission from Advanced Materials, Kirchhoff et al., 2026, © Wiley *[9]*.

Modeling Residual Stresses in OATMEAL

A natural concern when introducing an amorphous interlayer at every ply interface is how it will affect residual stresses throughout the laminate. To evaluate this, cross-ply $[0_2/90_2]$ panels were fabricated using both OATMEAL and neat CF/PEEK within the same vacuum bag to ensure

identical processing conditions. In this configuration, the final curvature of the panels serves as a direct "sensor" of the residual stresses generated during manufacturing.

Laminates were consolidated at 300°C and 380°C for 30 minutes, followed by controlled cooling at 5°C/min. Upon release from the tool, the resulting free strains caused the panels to adopt bi-stable cylindrical shapes. As expected, the CF/PEEK laminate processed at 300°C did not consolidate, as this temperature is below the melting temperature of the matrix. Surface curvature was quantified using point clouds acquired with a Keyence VL-800 optical profilometer. As shown in Figure 4, both OATMEAL panels are significantly flatter than the neat CF/PEEK reference, exhibiting approximately **30% lower curvature**, and notably showing minimal sensitivity to processing temperature.

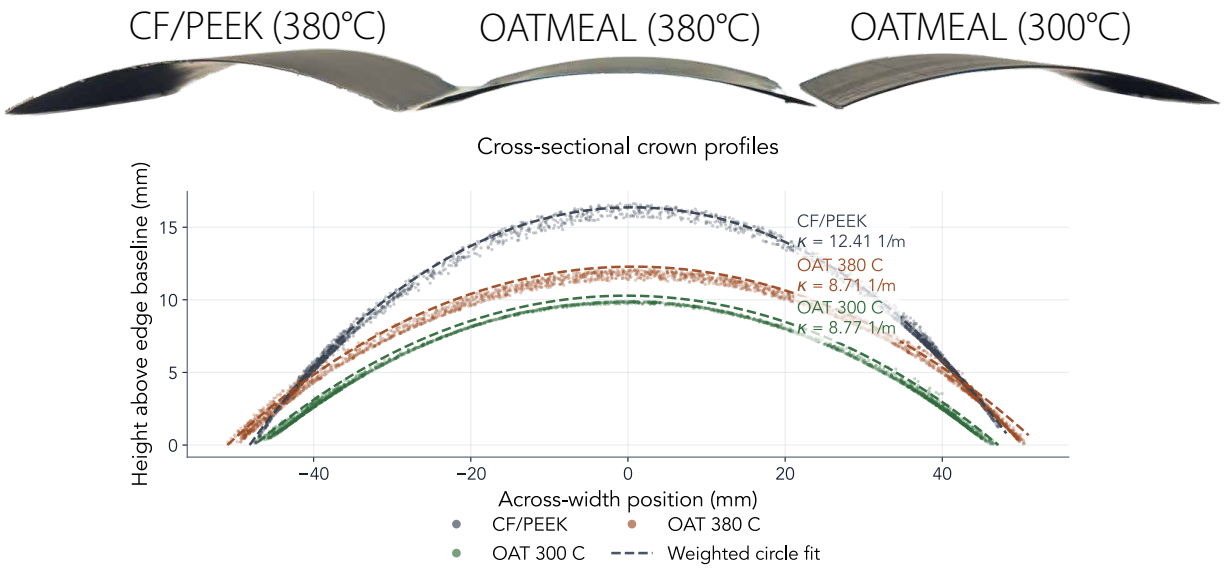

Figure 4. Processing induced warpage of $[0_2/90_2]$ cross-ply laminates provide insight into residual stresses. A Keyence VL-800 is used to extract the curvature information of the panels.

To understand the origin of this reduced curvature, a simple classical laminate theory (CLT) framework was used to compute laminate-scale thermal force resultants, curvature, and bulk membrane stress. Rather than explicitly modeling the full thermal and crystallization history, the analysis adopts a constant property thermoelastic formulation in which the effects of processing are condensed into a single effective stress-free temperature, T_{sf} [14]. This approach enables a direct comparison between material systems while isolating the role of interlaminar behavior.

The model assumes a small-strain, linear-elastic thermoelastic response during cooling. The CF/PEEK plies are treated as orthotropic lamina under plane-stress conditions, while the PEI-rich interlayers are modeled as isotropic. Perfect bonding between layers is assumed, and each layer is assigned homogeneous, temperature-independent properties. Within each scenario, residual stresses arise solely from thermal expansion mismatch; crystallization-induced shrinkage and other path-dependent effects are not explicitly included.

Material properties for the semi-crystalline plies were taken to be representative of AS4/APC-2 CF/PEEK ($E_1 = 131$ GPa, $E_2 = 8.7$ GPa, $G_{12} = 5.0$ GPa, $\nu_{12} = 0.28$, $\alpha_1 = 0.4 \times 10^{-6}$ /°C, $\alpha_2 = 30 \times 10^{-6}$ /°C) [9], [13]. The PEI-rich layers were modeled as isotropic Ultem 1000 with $E = 3.0$ GPa, $\nu = 0.36$, and $\alpha = 57 \times 10^{-6}$ /°C. A constant CF/PEEK thickness of 140 μm was assumed throughout the laminate along with 10 μm thick PEI interfaces. The PEI interlayers were modeled as discrete lamina with isotropic properties. In the analysis, the final temperature was taken to be 25°C.

A key outcome of this analysis is that the simplified CLT framework accurately predicts the warpage of OATMEAL laminates despite neglecting both explicit crystallization physics and temperature-dependent material properties. When the effective stress-free temperature is taken near the PEI glass transition temperature, approximately 217 °C, the predicted curvature agrees with experiment to within 2–3% (Figure 5). In contrast, the same simplified modeling approach is less accurate for conventional CF/PEEK laminates. For CF/PEEK, a higher effective stress-free temperature of approximately 270 °C provides a better approximation, consistent with stress development after crystallization and solidification, but still results in an error of approximately 13%. This discrepancy suggests that accurate prediction of conventional CF/PEEK warpage requires additional path-dependent physics, including crystallization kinetics, crystallization-associated shrinkage, temperature-dependent modulus evolution, temperature-dependent thermal expansion, and stress relaxation [6].

Mechanistically, this difference reflects the temperature range over which each laminate becomes mechanically constrained during cooling. In conventional CF/PEEK, residual stress begins to develop as the PEEK matrix crystallizes, stiffens, and loses its ability to relax deformation. During this same temperature window, the matrix also undergoes thermal contraction and crystallization-associated densification. These effects are therefore coupled: the magnitude of the residual stress depends not only on the amount of shrinkage, but also on when that shrinkage occurs relative to the evolving matrix modulus, thermal expansion behavior, and relaxation time scale.

A simple scale estimate illustrates why crystallization-associated shrinkage cannot be ignored for CF/PEEK. Crystallization of initially amorphous PEEK is commonly associated with a volumetric shrinkage of several percent [15]. In a unidirectional composite ply, only the matrix phase contributes directly to this shrinkage, so the effective ply-level transverse shrinkage scales with the matrix volume fraction, approximately $1 - V_f$. For a typical fiber volume fraction near 0.6, this places the composite-scale shrinkage strain in a range large enough to influence curvature in cross-ply laminates, where transverse contraction of each ply is constrained by neighboring plies of different orientation. However, this estimate should be interpreted as solely a component of the overall warpage. Crystallization shrinkage alone does not determine the residual stress state; instead, warpage arises from the coupled evolution of crystallinity, matrix modulus, thermal expansion, thermal contraction, and stress relaxation during cooling [15].

This coupling explains why a single stress-free temperature is only an approximation for conventional CF/PEEK. The laminate is not stress-free above one defined temperature and fully constrained below it. Instead, mechanical constraint develops progressively as the matrix crystallizes, stiffens, and changes thermal expansion behavior. The effective stress-free

temperature of approximately 270 °C therefore represents an empirical collapse of a distributed, path-dependent process into a single thermoelastic parameter. The remaining curvature error is consistent with the simplified CLT model neglecting the underlying crystallization kinetics, modulus evolution, crystallization-associated shrinkage, temperature-dependent thermal expansion, and relaxation behavior.

OATMEAL changes this stress-development process by introducing PEI-rich interlayers between the CF/PEEK plies. Above the PEI glass transition, these interlayers remain rubbery and relatively compliant. As a result, thermal strains and crystallization-associated shrinkage that develop in the CF/PEEK plies between the PEEK crystallization regime and the PEI glass transition are not fully transferred into laminate-scale residual stress. Only as the PEI-rich interlayers stiffen near 217 °C do these strains become strongly constrained. This delayed mechanical lock-in explains why OATMEAL warpage can be captured using a single effective stress-free temperature near the PEI glass transition, even when the model uses temperature-independent material properties, whereas conventional CF/PEEK requires a more complete crystallization-aware and temperature-dependent process model.

This delayed stress lock-in has two important consequences. First, it reduces the effective residual-stress magnitude, consistent with the observed decrease in curvature. Second, it makes the laminate response less sensitive to the peak processing temperature, 300 °C versus 380 °C, because the dominant constraint is set by PEI Tg rather than by the detailed crystallization history of the PEEK matrix. The key point is that OATMEAL does not simply reduce warpage by changing material properties; it changes when the laminate becomes mechanically constrained. By shifting stress lock-in from the PEEK crystallization regime to the PEI glass transition, OATMEAL converts a coupled crystallization-dependent, temperature-dependent residual-stress problem into one that can be approximated with a much simpler constant-property thermoelastic framework.

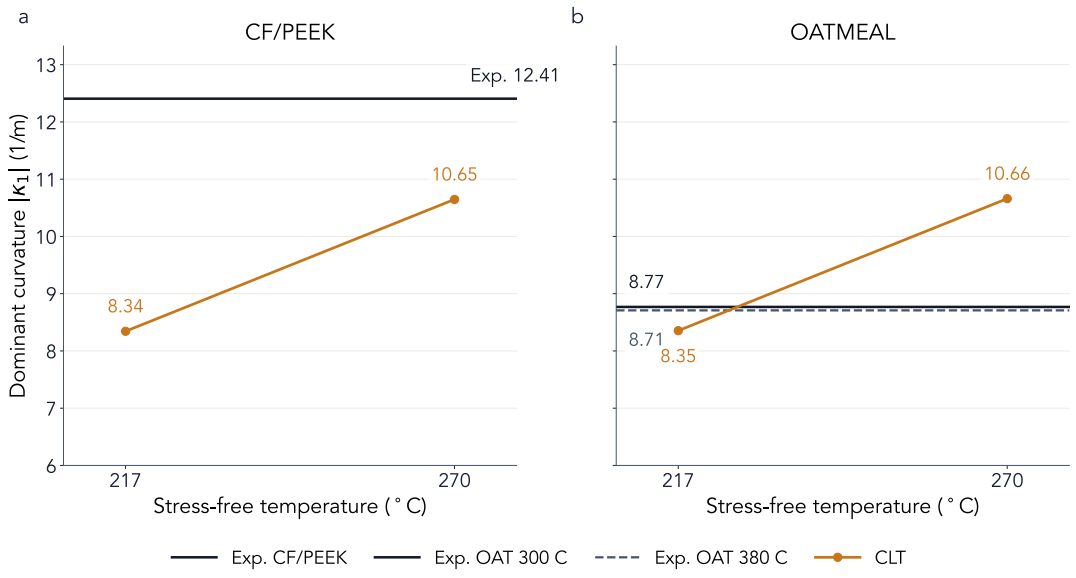

Figure 5. Measured and CLT-predicted curvature for the (a) CF/PEEK and (b) OATMEAL cross-ply laminates.

Why Does This Matter?

For OATMEAL, global laminate behavior can be accurately captured using CLT with constant properties and a single effective stress-free temperature. In contrast, conventional CF/PEEK requires additional crystallization physics to achieve comparable agreement. By shifting the effective stress lock-in temperature toward the PEI glass transition and decoupling crystallization shrinkage from the interlaminar response, OATMEAL reduces both the magnitude and complexity of the residual stress state. As summarized in Table 1, this removes the need to model crystallization behavior.

The practical impact is substantial. By collapsing a multi-physics, path-dependent problem into a simplified thermoelastic framework, OATMEAL reduces material characterization requirements, lowers computational cost, and enables faster design iteration. More importantly, it provides a predictable and robust foundation for process development, where laminate behavior can be reliably anticipated without resolving the full complexity of crystallization-driven physics.

Table 1. Governing physics: Conventional CF/PEEK vs. OATMEAL

Physics	Conventional (CF/PEEK)	OATMEAL	What this means
Crystallization	Governs cooling and stress development	Removed from bonding step	No need to model crystallization kinetics
Crystallization Shrinkage	Crystallization-induced, path-dependent	Not active during bonding	Residual stress no longer path-dependent
Material state during bonding	Melt → crystallizing solid	Solid + rubbery interface	Properties remain stable during joining
Stress lock-in	Near crystallization (~270 °C)	At PEI Tg (~217°C)	Single temperature controls residual stress
Thermal history sensitivity	High	Low	Can be captured with one (T_{sf})

Part Consolidation: 5x increase in throughput with 75% less energy

So, why go through the effort of engineering fully crystallized prepreg with thin amorphous surfaces for bonding? The premise is simple: ***if you don't melt your crystals, you don't have to regrow them and you are no longer constrained upon cooling.***

Imagine an oven held at temperature while parts are continuously loaded and unloaded, enabling a continuous-flow process (see figure 6). In this paradigm, throughput is no longer governed by cooling constraints, but by the time required for interfacial healing. A simple thermal model predicts a **5× increase in throughput along with 75% reduction in energy (from not cycling the oven)** under conservative assumptions, with further gains possible through multi-part handling or rapid-entry heating (e.g., IR-assisted processing) [9]. In the limit, cycle time approaches only the dwell time needed for PEI-PEI healing at interfaces. To demonstrate this, a VBO process was evaluated, a scalable approach not limited by press or autoclave size, where

OATMEAL and hybrid laminates were consolidated at 300°C under vacuum and allowed to cool freely in air. Hybrid laminates are the PEI-PEEK prepreg prior to laser ablation. Measured cooling rates reached ~45°C/min, nearly an order of magnitude faster than the controlled 5°C/min rates typically required for crystallization-driven processing.

Although both hybrid and OATMEAL laminates achieved full PEEK crystallinity under fast-cooled VBO conditions, their mechanical responses diverged significantly. The hybrid laminate exhibited a substantial drop in short-beam shear strength (SBS), decreasing from 88 ± 4 MPa (autoclave) to 41 MPa (VBO), a 53% reduction, likely due to the smooth PEI film surface preventing interlaminar void removal under low pressure.

In contrast, OATMEAL maintained high performance, with SBS decreasing only modestly from 96 ± 4 MPa to 82 ± 8 MPa. This improved robustness is attributed to two key features: (i) reduced resin-rich interlayers, and (ii) engineered interlaminar surfaces that promote effective bonding even under rapid thermal cycles. As a result, OATMEAL delivers a **~98% improvement in SBS relative to the hybrid laminate under fast-cooling conditions**, while enabling dramatically shorter cycle times.

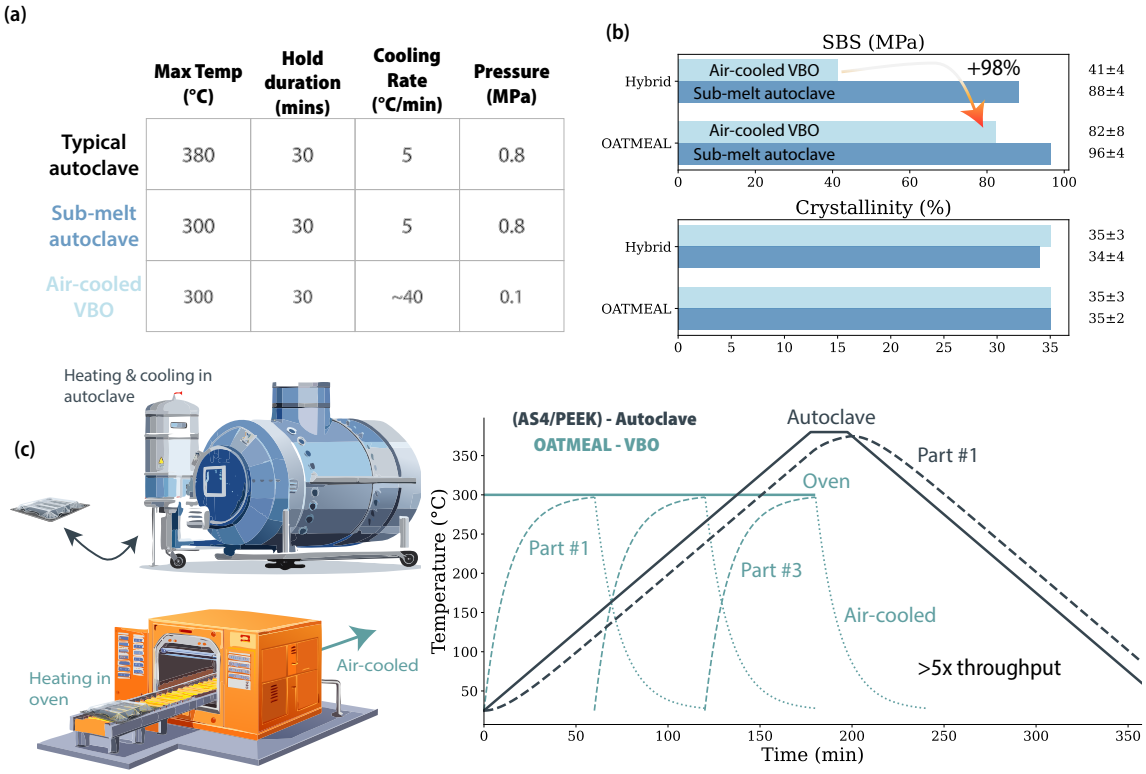

Figure 6. SBS performance for OATMEAL vs. hybrid (non-ablated PEI-PEEK prepreg) for the fast-VBO process compared to typical autoclave along with demonstration of constant temperature oven with parts shuffling in/out showing 5x throughput increase. Reproduced with permission from Advanced Materials, Kirchhoff et al., 2026, © Wiley [9].

An additional important outcome of this study is the emergence of a processing window in which the semi-crystalline portion remains compliant. Within this Goldilocks zone (~217–300°C), the PEI interlayers are in a rubbery state while the PEEK matrix remains semi-crystalline and

dimensionally stable. As a result, interlaminar shear stiffness is reduced, allowing plies to deform without requiring full melting of the matrix. This suggests that OATMEAL laminates may be thermoformed below the melt temperature, with deformation accommodated by compliant interlaminar regions rather than bulk flow. Forming may occur while the laminate is compliant, bonding is governed by interfacial healing, and residual stresses are only locked in upon T_g of the PEI. This represents a departure from conventional thermoplastic processing, where melting, forming, and crystallization are tightly coupled.

Notably, performance is governed by the retained PEI content and its impact on local fiber volume fraction. In the present study, the fiber volume fraction decreases from 59.6 ± 2.1% in the baseline CF/PEEK prepreg to 38.1 ± 3.4% after adding 25 μm PEI and recovers to 45.9 ± 2.0% following laser ablation. Ongoing efforts at NASA Langley are focused on optimizing PEI–PEEK interdiffusion and refining laser ablation strategies to further balance interfacial bonding with structural efficiency. This highlights the central trade-off in OATMEAL: maintaining sufficient amorphous material for robust interfacial bonding while preserving fiber volume fraction and load-carrying capability.

CONCLUSIONS

This work demonstrates that amorphous bonding provides a fundamentally different pathway for joining and manufacturing thermoplastic composites, one that decouples interfacial healing from crystallization. Using Thermabond® as a controlled model problem, bond performance was shown to be governed primarily by processing (i.e., sufficient interdiffusion), and highlighted key lessons like PEEK being a better semi-crystalline polymer to pair with PEI as well as the challenges of handling thin PEI films. These results establish that thin amorphous interlayers can enable sub-melt joining without relying on melt-driven crystallization.

Extending this concept to laminate-scale manufacturing, the OATMEAL prepreg architecture enables amorphous bonding for laminate consolidation. Cross-ply warpage experiments reveal that, contrary to expectations, repeated amorphous interfaces do not amplify residual stress. Instead, they reduce cross-ply laminate curvature (reduced by 30%) by lowering the effective stress lock-in temperature and eliminating crystallization-induced shrinkage from the interlaminar response. As a result, laminate behavior can be accurately predicted using classical laminate theory with a single effective stress-free temperature, enabling predictability to a process that is otherwise strongly path-dependent in conventional CF/PEEK systems due to crystallization.

From a manufacturing perspective, OATMEAL enables a shift from crystallization-limited processing to healing-limited processing. By preserving crystallinity during consolidation, it removes the need for controlled cooling and allows rapid thermal cycles without sacrificing performance. In a fast-cooled VBO process designed for increase in throughput, OATMEAL laminates maintain aerospace-grade short beam strength under air cooling. This decoupling of bonding from crystallization enables continuous-flow manufacturing concepts and is projected to increase throughput by up to fivefold while reducing energy consumption by approximately 75% by removing the need to cycle oven temperatures.

Taken together, these results establish amorphous bonding not only as a viable joining strategy, but as a scalable manufacturing paradigm for high-performance thermoplastic composite parts. By simplifying both the governing physics and the processing requirements, OATMEAL provides a pathway toward more predictable, energy-efficient, and high-rate composite manufacturing aligned with the demands of next-generation aerospace systems.

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