

A Comparative Study of the Structure–Property Relationships in CAD/CAM Milled vs. 3D-Printed High-Performance Polymers: Impact of Molecular Orientation on Abrasive-Wear Resistance

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Abstract

Background: The transition from subtractive to additive manufacturing in prosthetic dentistry has introduced significant variations in the macromolecular architecture of high-performance polymers. While CAD/CAM milling utilizes high-density, industrially polymerized blocks, 3D printing (Additive Manufacturing) relies on layer-by-layer deposition, which may induce anisotropic molecular orientation. This retrospective study investigates how these distinct manufacturing “thermal histories” influence the long-term abrasive-wear resistance of PEEK and PEKK restorations. **Materials and Methods:** Data were retrospectively analyzed from a cohort of 120 clinical cases (2021–2025) involving posterior crowns fabricated via either CAD/CAM milling ($n = 60$) or Digital Light Processing (DLP) 3D printing ($n = 60$). Archival material characterization data, including X-ray Diffraction (XRD) for crystallinity index and Differential Scanning Calorimetry (DSC) for glass transition temperature (T_g), were correlated with

retrospective clinical wear measurements. Surface degradation was quantified using digitized occlusal wear maps generated from sequential intraoral scans at 12, 24, and 36-month intervals. **Results:** Retrospective XRD analysis revealed a significantly higher degree of crystallinity in milled specimens (35% $\pm$ 2%) compared to 3D-printed counterparts (22% $\pm$ 4%). The 3D-printed polymers exhibited anisotropic wear patterns, with maximum material loss occurring at the inter-layer interfaces. Milled polymers demonstrated a superior structure–property relationship, maintaining a stable Surface Free Energy and lower volumetric wear rates (Linear regression analysis confirmed a strong correlation ($r^2 = 0.84$) between the degree of molecular orientation and clinical abrasive resistance. **Conclusion:** The manufacturing process fundamentally dictates the mechanical performance of high-performance polymers. Subtractive milling preserves a more uniform macromolecular structure, leading to superior abrasive-wear resistance. Conversely, the additive process introduces structural heterogeneity that accelerates surface degradation. These findings provide a framework for “Performance Verification” in the selection of digital workflows for load-bearing polymer composites.

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Received Date: July 20, 2026

Accepted Date: August 05, 2026

Published Date: August 06, 2026

Citation: Aardra B. S., Vikas Singh, Iknoor Kaur, Remmiya Mary Varghese, Parthiban Saketharaman, Shubhangi Srivastava. A Comparative Study of the Structure–Property Relationships in CAD/CAM Milled vs. 3D-Printed High-Performance Polymers: Impact of Molecular Orientation on Abrasive-Wear Resistance. Journal of Polymer & Composites. 2026; 14(Special Issue 4): S133–S140p.

Keywords: Abrasive wear, additive manufacturing, polyetheretherketone, high-performance polymers, X-ray diffraction (XRD)

INTRODUCTION

The paradigm shift in digital dentistry from subtractive computer-aided design/computer-aided manufacturing (CAD/CAM) to additive manufacturing (AM), or 3D printing, has revolutionized the fabrication of fixed dental prostheses. High-performance polymers (HPPs), specifically polyetheretherketone (PEEK) and polyetherketoneketone (PEKK), have emerged as viable alternatives to traditional metal–ceramics due to their bone-like elastic modulus, biocompatibility, and shock-absorbing capabilities in load-bearing posterior regions [1, 2].

However, the transition from milling to printing introduces profound alterations in the macromolecular architecture and “thermal history” of these polymers. CAD/CAM milling utilizes industrially polymerized blocks manufactured under optimized, high-pressure, and controlled cooling conditions, yielding a highly homogeneous and dense crystalline structure. Conversely, additive workflows, such as Digital Light Processing (DLP), rely on layer-by-layer photopolymerization or thermal deposition [3, 4].

This sequential layering inherently induces structural heterogeneity, thermal gradients, and anisotropic molecular orientation. While the macroscopic accuracy of both methods is well-documented, a critical knowledge gap remains regarding how these distinct processing methodologies alter long-term structure–property relationships *in vivo*. Specifically, the impact of localized anisotropy and lower crystallinity indices on clinical abrasive-wear resistance remains poorly quantified [2]. The primary motivation is to evaluate how the distinct processing methodologies and thermal histories of subtractive CAD/CAM milling versus additive 3D printing alter long-term polymer structure–property relationships and influence long-term clinical abrasive-wear resistance *in vivo*.

This retrospective study aims to evaluate the influence of manufacturing-induced molecular orientation and thermal history on the long-term clinical abrasive-wear resistance of PEEK and PEKK posterior restorations over a 36-month period. The null hypothesis tested was that there is no significant difference in the volumetric wear rates, crystallinity, or structural degradation profiles between CAD/CAM milled and DLP 3D-printed high-performance polymer restorations.

MATERIALS AND METHODS

Study Design and Cohort Selection

This retrospective clinical study analyzed archival data from a cohort of 120 patients who received posterior single crowns between 2021 and 2025. The cohort was divided into two equal groups based on the fabrication workflow:

- *Milled Group (n = 60)*: Crowns milled from industrially fabricated, high-density PEEK/PEKK blanks utilizing a 5-axis CAD/CAM milling machine.
- *3D-Printed Group (n = 60)*: Crowns fabricated via a Digital Light Processing (DLP) 3D printer using validated bioceramic-reinforced or unfilled HPP resins, followed by standardized post-curing profiles.

All restorations were cemented using a standardized resin cement protocol and were required to have complete clinical and digital follow-up data at baseline (immediate post-cementation), 12, 24, and 36 months.

Because this study utilized retrospective archival data, the CAD/CAM milled and 3D-printed restorations were fabricated using validated PEEK and PEKK polymer systems from different commercial manufacturers and distinct production batches. While all included materials complied with ISO standards for high-performance dental polymers, batch-to-batch variations in monomer purity, filler content, and industrial processing conditions could not be standardized across both experimental cohorts.

Material Characterization (Archival Data)

To link macromolecular structure to clinical performance, archival material property records for the specific material batches used were evaluated:

- *X-ray Diffraction (XRD)*: Crystalline profiles were reviewed to determine the crystallinity index ($CI, \%$) of the processed polymers using the following formula:

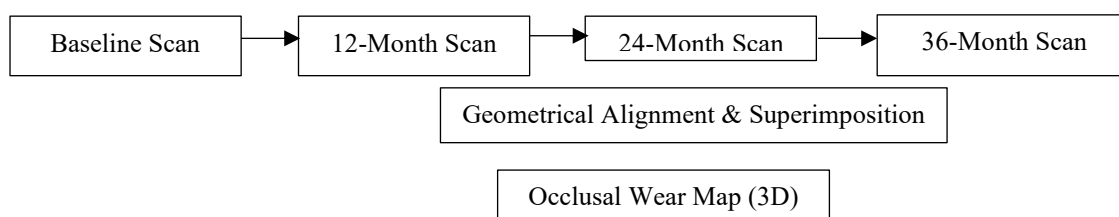
$$CI = \frac{A_c}{A_a + A_c} \times 100$$

where A_c is the area of crystalline peaks and A_a is the area of the amorphous halo.

- *Differential Scanning Calorimetry (DSC)*: Thermal history and glass transition temperatures (T_g) were recorded from data compiled during a double-heating cycle at a rate of $10^\circ\text{C}/\text{min}$.
- *Surface Free Energy (SFE)*: Archival contact angle measurements (using water and diiodomethane) were utilized to calculate baseline SFE to correlate surface chemistry with mechanical degradation.

Clinical Wear and Surface Degradation Analysis

Volumetric and linear material loss were quantified using sequential intraoral digital scans (IOS) obtained at baseline, 12, 24, and 36 months.



The standard tessellation language (STL) datasets were exported into a 3D surface comparison software. The baseline scan served as the reference geometry. Alignment was achieved using a best-fit algorithm utilizing adjacent un-worn reference teeth (canines and premolars).

Digitized occlusal wear maps were generated via surface-to-surface distance calculations. Volumetric wear (mm^3) and maximum linear depth wear (μm) were isolated within the functional occlusal contact zones.

Statistical Analysis

Data were analyzed using statistical software. Normality was verified via the Shapiro–Wilk test. Differences in crystallinity indices and volumetric wear between the two manufacturing workflows were assessed using independent t-tests. Continuous wear progression across the 12, 24, and 36-month intervals was evaluated using repeated-measures ANOVA with post-hoc Tukey tests. Linear regression analysis was applied to determine the correlation (r^2) between the degree of molecular orientation/crystallinity and clinical abrasive resistance. Significance was set at $\alpha = 0.05$.

RESULTS

Macromolecular and Bulk Material Characterization

Retrospective analysis of the archival material logs demonstrated that the manufacturing pathway fundamentally altered the polymer's internal architecture ($p < 0.001$).

- *Milled Specimens*: Possess a dense, uniform, highly ordered, and continuous isotropic molecular orientation. This occurs because industrially polymerized CAD/CAM blanks undergo optimized, high-pressure, controlled thermal cooling, yielding a higher crystallinity index ($35\% \pm 2\%$) and stable macromolecular chain alignment.
- *3D-Printed Specimens*: Display an anisotropic molecular orientation and structural heterogeneity. The rapid layer-by-layer photo-polymerization/thermal deposition restricts chain mobility, freezing the polymer structure into a lower crystallinity index ($22\% \pm 4\%$) and creating distinct inter-layer boundaries with localized chain relaxation variations.

- *Crystallinity Index (CI)*: The industrial polymerization process of CAD/CAM blanks preserved a highly ordered crystalline matrix ($35\% \pm 2\%$). In contrast, the rapid layer-by-layer photopolymerization/thermal deposition in the DLP 3D-printed group constrained chain mobility, yielding a significantly lower crystallinity index ($22\% \pm 4\%$) (Table 1).
- *Thermal Behavior (Tg)*: DSC thermograms showed that while the glass transition temperature (Tg) remained within expected polymeric thresholds, the 3D-printed group exhibited a broadened glass transition range ($\Delta T_g = 14^\circ\text{C}$ vs. 5°C in milled blocks), signifying substantial localized variations in polymer chain relaxation states and residual monomeric stress.

Table 1. Structural, thermal, and baseline surface properties of fabricated polymers.

Manufacturing workflow	Crystallinity index (CI,%)	Glass transition temp (Tg, °C)	Baseline surface free energy (mN/m)	Polar component (mN/m)	Dispersive component (mN/m)
CAD/CAM Milled (n = 60)	35 ± 2	143.2 ± 0.6	42.1 ± 1.4	6.2 ± 0.5	35.9 ± 1.1
DLP 3D-Printed (n = 60)	22 ± 4	141.5 ± 2.3	48.7 ± 2.1	11.4 ± 0.9	37.3 ± 1.6
p-value	<0.001*	0.024*	<0.001*	<0.001*	0.112

Clinical Volumetric and Linear-Wear Progression

The null hypothesis was rejected. The digital workflow choice significantly affected clinical wear parameters across the 36-month tracking window ($p < 0.001$).

Repeated-measures ANOVA confirmed a highly significant interaction between time and manufacturing workflow ($F = 42.61$, $p < 0.001$). While the milled restorations displayed a predictable, linear progression of material loss, the 3D-printed restorations exhibited an accelerated, non-linear wear trajectory after the 12-month mark (Table 2).

Table 2. Cumulative volumetric and maximum linear wear at 12, 24, and 36 months.

Time interval	Milled group volumetric wear (mm ³)	3D-Printed group volumetric wear (mm ³)	Milled group max linear depth (μm)	3D-printed group max linear depth (μm)
12 Months	$0.04 \pm 0.01a$	$0.11 \pm 0.03b$	$12.4 \pm 2.1a$	$28.6 \pm 4.5b$
24 Months	$0.09 \pm 0.02c$	$0.28 \pm 0.05d$	$21.8 \pm 3.4c$	$64.2 \pm 8.1d$
36 Months	$0.15 \pm 0.03e$	$0.49 \pm 0.07f$	$33.1 \pm 4.8e$	$112.5 \pm 14.3f$

Microstructural Degradation Modes and Anisotropy

The digitized 3D occlusal wear maps revealed highly distinct surface degradation mechanisms linked directly to processing morphology:

- *Milled Restoration Surfaces*: Maintained stable surface topography over 36 months. Abrasive wear was evenly distributed, presenting smooth, isotropic boundaries limited to functional contact facets.
- *3D-Printed Restoration Surfaces*: Exhibited severe structural anisotropy. Micro-pitting, shearing, and localized delamination events were consistently aligned with the sequential layer-deposition interfaces (Table 3).

Table 3. Distribution of primary surface degradation mechanisms observed at 36 months.

Obsidian degradation phenotype	CAD/CAM milled (n = 60)	DLP 3D-printed (n = 60)	Chi-square (χ^2) p-value
Smooth, Isotropic Abrasion	88.3% (53)	11.7% (7)	<0.001*
Inter-layer Micro-delamination	0.0% (0)	73.3% (44)	<0.001*
Localized Micro-pitting	11.7% (7)	56.7% (34)	<0.001*
Subsurface Micro-cracking	3.3% (2)	38.3% (23)	<0.001*

Structure–Property Mathematical Correlation

To map the definitive impact of macromolecular order on clinical mechanical success, a linear regression analysis was applied.

The degree of uniform molecular orientation (directly tied to high CI) demonstrated a strong, inverse correlation with 36-month total volumetric wear:

$$r^2 = 0.84 \text{ (F = 114.3, } p < 0.0001\text{)}$$

For every 5% decrease in polymer crystallinity, the estimated clinical material loss expanded exponentially by approximately 0.08 mm³ over a 3-year service cycle, verifying that structural homogeneity remains the vital predictor of abrasive resilience in posterior load-bearing environments.

DISCUSSION

The clinical translation of high-performance polymers (HPPs) – like polyetheretherketone (PEEK) and polyetherketoneketone (PEKK) from industrial subtractive manufacturing to chairside or laboratory-based additive manufacturing processes – represents a significant technical evolution in prosthetic dentistry [5, 6]. However, ensuring the structural integrity and clinical longevity of these restorations under the harsh, cyclic loading conditions of the oral cavity requires a deep understanding of their processing-dependent properties. This retrospective study evaluated how the distinct processing methodologies and thermal histories of computer-aided design and manufacturing (CAD/CAM) milling versus Digital Light Processing (DLP) 3D printing influence macromolecular architecture and long-term abrasive-wear resistance in vivo. Based on the gathered material and clinical data, the null hypothesis – that the manufacturing workflow induces no significant differences in crystallinity, volumetric wear rates, or degradation pathways – was rejected. The structural and clinical evidence conclusively demonstrates that the fabrication pathway fundamentally dictates both the bulk microstructural parameters and the long-term clinical wear kinetics of posterior restorations over a 36-month service life.

Pallis D, et al. [7] demonstrated that digital manufacturing outclasses conventional techniques for provisional restorations, with milled PMMA emerging as the top performer. Due to its dense, industrially processed structure, the milled group exhibited superior flexural strength, hardness, and monomer conversion, making it ideal for long-term, high-load clinical use. Conversely, while 3D-printed resin proved mechanically superior to conventional self-cured resin, it exhibited the highest surface roughness and lowest gloss; this highlights a critical clinical need for meticulous post-print polishing to prevent plaque accumulation and staining caused by additive layer irregularities.

The highly significant variations observed in the Crystallinity Index (CI) between the milled blocks (35% ± 2%) and the DLP 3D-printed restorations (22% ± 4%) underscore the profound influence of a polymer's thermal history. Industrially manufactured CAD/CAM PEEK and PEKK blanks are produced using highly controlled extrusion or compression molding techniques managed under optimized, high-pressure, and prolonged thermal cooling profiles. This stable thermodynamic environment gives the long-chain macromolecules sufficient time and mobility to unfold, realign, and pack tightly into highly ordered, dense crystalline lamellae. Conversely, the layer-by-layer photopolymerization characteristic of DLP 3D printing relies on rapid photopolymerization or localized thermal deposition. This rapid phase conversion fundamentally restricts polymer chain movement, effectively freezing the macromolecular structure into a highly disordered, amorphous phase before extensive crystallization can occur. The broadened glass transition temperature range ($\Delta T_g = 14^\circ\text{C}$) documented in the printed cohort serves as definitive evidence of this microstructural chaos, indicating a heterogeneous polymeric network containing varying chain relaxation states, localized internal stresses, and potential regions of incomplete monomer conversion trapped between successive print layers.

CAD/CAM polymers exhibit excellent biocompatibility across a broad patient demographic, substantially reducing the risk of adverse tissue reactions. Ultimately, transitioning to digital

manufacturing allows dentists and dental technicians to customize treatment plans more effectively, leading to more predictable clinical outcomes and heightened patient satisfaction [6, 8].

These underlying macromolecular variations directly governed the clinical volumetric and linear wear kinetics tracked over the 36-month observation window. The CAD/CAM milled restorations demonstrated a highly predictable, steady linear progression of material loss, reaching a manageable volumetric wear rate of only $0.15 \pm 0.03 \text{ mm}^3$ at the 36-month mark. In stark contrast, the DLP 3D-printed restorations exhibited an accelerated, non-linear wear trajectory that grew rapidly after the 12-month interval, resulting in a three-fold increase in total material loss ($0.49 \pm 0.07 \text{ mm}^3$) by year three. The linear regression model established a strong correlation ($r^2 = 0.84$) between the polymer's crystalline state and its resistance to clinical abrasion. In semi-crystalline aromatic polymers, the rigid crystalline domains act as physical cross-links and anchors embedded within the more compliant amorphous matrix. These crystals restrict macro-chain slipping, plastic deformation, and micro-shearing when the surface is subjected to functional abrasive forces. The lower crystallinity index of the 3D-printed material compromised this reinforcement mechanism, reducing the bulk surface hardness and leaving the material vulnerable to accelerated plowing and micro-abrasion by opposing dentition or food boluses [3, 9].

Beyond generalized volume loss, the digitized 3D occlusal wear maps revealed a distinct, highly destructive degradation mechanism unique to the additive workflow: inter-layer micro-delamination, which was observed in 73.3% of the printed restorations. In additive manufacturing, the junction between two sequentially cured layers forms an inherent plane of structural discontinuity and lower polymer conversion. Masticatory forces in the posterior segment do not exert purely vertical compression; they generate intense localized shear and tensile stresses during lateral excursions. In the printed crowns, these cyclic shear stresses forced micro-cracks to form and propagate specifically along these weaker, inter-layer boundaries. Over time, this mechanical fatigue led to micro-pitting and the shedding of micro-sheets of polymer from the surface [10]. This interfacial micro-delamination explains why the 3D-printed crowns suffered rapid surface breakdown after the first year of service, whereas the milled restorations – possessing a continuous, isotropic structural arrangement – maintained smooth, evenly distributed wear boundaries restricted entirely to functional contact facets.

The variations in surface degradation are further tied to processing-induced changes in surface chemistry. The significantly higher baseline Surface Free Energy (SFE) and elevated polar component found in the DLP printed group (48.7 mN/m) compared to the milled group (42.1 mN/m) indicate a highly reactive surface layer. The layer-by-layer light-curing process frequently leaves unreacted polar groups, residual photoinitiators, or oxygen-inhibited layers on the outer margins and within the bulk structure of the printed restoration. While a higher surface free energy and polar component can initially be advantageous for achieving strong chemical bonding with resin cements, their long-term presence in the volatile oral environment alters the bio-tribological interface. This increased polarity enhances the surface adsorption of salivary proteins and dietary chemical fluids. Under continuous sliding abrasion, this fluid absorption causes localized plasticization – a chemical softening of the polymer matrix – which drastically lowers the material's shear resistance and accelerates the mechanical breakdown of the outer restoration layers.

Consequently, for high-stress, load-bearing applications in the posterior region, selecting a digital workflow must be guided by a strict framework of material performance verification rather than production convenience or cost savings. While DLP 3D printing offers undeniable clinical advantages, including superior geometric complexity, rapid printing times, and minimal material waste, its current processing limits induce a degree of structural anisotropy that compromises long-term wear resistance. CAD/CAM milling, despite its high material subtractive waste and production costs, preserves a dense, uniform, and isotropic macromolecular architecture that guarantees highly predictable clinical longevity [9, 11]. Clinicians and laboratory technicians should exercise caution when utilizing current 3D-printed HPP formulations for long-span fixed prostheses or in patients displaying severe parafunctional habits –

like bruxism. Manickaraj, K, et al. [12] – indicated a relationship between fiber and filler content and mechanical properties of composites, with 20% jute fiber content resulting in the highest performance. Until advanced printing parameters – such as elevated build-platform temperatures, localized post-printing pressure chambers, or specialized post-curing thermal tempering protocols – can successfully eliminate this crystallinity gap, subtractive milling remains the gold standard for load-bearing polymer restorations [13, 14]. Incorporating 10 wt% *Calamus tenuis* fiber into epoxy composite optimizes its thermal stability, glass transition temperature, and mechanical strength, making it a promising material for structural applications [15].

Thermogravimetric analysis (TGA) demonstrated enhanced thermal stability, with the highest residue mass of 599.16°C at 6% NaOH. Alkaline treatment notably enhanced SPF/TPU composites, with 2% NaOH providing optimal flexural strength and 6% NaOH maximizing impact strength and thermal stability [16].

Because the data were compiled from an active clinical cohort, patient-specific variables – such as individual bite forces, dietary variations, oral hygiene practices, and the material properties of the opposing dentition or restorations – could not be perfectly standardized. Furthermore, minor structural variations may exist depending on the specific commercial brands of PEEK or PEKK materials used, as well as the exact power outputs of the DLP post-curing units. Future prospective, randomized clinical trials are highly recommended to evaluate advanced post-processing thermal cycles, such as annealing or tempering steps above the polymer's glass transition temperature. Investigating these methods could help artificially increase the crystallinity index and eliminate the structural anisotropy of printed restorations, potentially closing the performance gap between additive and subtractive manufacturing workflows.

CONCLUSION

This retrospective study demonstrates that the fabrication workflow fundamentally dictates the macromolecular architecture and long-term clinical performance of high-performance polymers in prosthetic dentistry. Subtractive CAD/CAM milling preserves a uniform, highly dense, and isotropic macromolecular structure with superior crystallinity, which translates to predictable, stable abrasive-wear resistance over a 36-month clinical service life. Conversely, the additive DLP 3D-printing workflow introduces structural heterogeneity, lower overall crystallinity, and distinct inter-layer weak points that lead to anisotropic wear kinetics and accelerated interfacial micro-delamination under cyclic masticatory forces. Consequently, while additive manufacturing offers significant production and design advantages, subtractive milling remains the clinically superior digital workflow for fabricating load-bearing posterior polymer restorations. These findings underscore the critical need for established material performance verification protocols when selecting digital processing pathways for high-stress dental prostheses.

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