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Effect of annealing on the room-temperature dry sliding tribological behavior of additively manufactured neat and graphene-reinforced PLA

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Abstract

The influence of annealing on the dry reciprocating tribological behavior of fused filament fabrication (FFF)-printed neat polylactic acid (PLA) and graphene-reinforced PLA was investigated. Specimens were annealed at 90, 100, and 120 °C for 60, 120, and 240 min and tested under dry reciprocating sliding against an Al₂O₃ ball according to ASTM G133. The effects of annealing on the coefficient of friction and specific wear coefficient were evaluated and correlated with crystallinity and mechanical properties of the same materials. Worn surfaces were examined by stereomicroscopy and scanning electron microscopy to identify the dominant wear mechanisms. Annealing significantly improved the wear resistance of both materials. Relative to the as-printed condition, the specific wear coefficient decreased by up to approximately 74% for neat PLA and 93% for graphene-reinforced PLA, while the friction coefficient increased by up to 43% and 11%, respectively, showing that reductions in wear were not necessarily accompanied by reductions in friction. The reduction in wear was correlated with annealing-induced changes in crystallinity and mechanical properties, with tensile and flexural strengths exhibiting the strongest relationships with the specific wear coefficient. Graphene-reinforced PLA consistently exhibited lower wear than neat PLA under all investigated conditions, suggesting that its superior wear resistance cannot be explained solely by the bulk mechanical properties considered in this study. Microscopy revealed a transition from extensive plastic deformation and delamination in neat PLA to predominantly mild microploughing, localized cracking, and compacted debris layers in graphene-reinforced PLA. Under the investigated dry reciprocating sliding conditions, the combination of annealing and graphene reinforcement effectively improved the wear resistance of FFF-printed PLA.

Keywords: Additive manufacturing; Fused filament fabrication; Dry sliding wear; Friction; Annealing; Graphene-reinforced PLA.

1. Introduction

Fused filament fabrication (FFF) has become a widely used additive manufacturing route for producing polylactic acid (PLA) components due to its low processing temperature, biodegradability, ease of printing, and cost-effectiveness. However, when neat PLA is used in sliding contacts, its tribological performance remains limited by its relatively low toughness, tendency to microcracking, and sensitivity to the structural features generated during layer-by-layer deposition [1–3]. Unlike conventionally processed polymers, FFF-printed PLA is composed of deposited roads, raster interfaces, interlayer boundaries, and residual voids. Therefore, its wear resistance is not governed only by the intrinsic properties of PLA, but also by interlayer bonding, surface roughness, printing orientation, load-bearing capacity, and transfer-film stability during sliding [1–6].

For neat PLA, several studies have shown that printing parameters strongly affect friction and wear. Layer thickness, build orientation, infill density, infill pattern, and extrusion temperature modify the real contact area, surface morphology, and mechanical integrity of the printed structure [1–6]. Zhiani Hervan et al. [1] reported that layer thickness and build orientation affect hardness, coefficient of friction, and wear rate of FDM-printed PLA. Hanon and Zsidai [2] also showed that printing orientation changes the wear depth and worn-surface morphology, confirming the anisotropic tribological behavior of printed PLA. These results indicate that the deposited filament architecture controls how the contact load is supported and how material removal occurs during sliding.

Extrusion temperature also plays an important role in the wear behavior of neat PLA. Zainal et al. [3] observed that increasing the extrusion temperature reduced the specific mass loss of 3D-printed PLA. This improvement was attributed to better filament coalescence, reduced interlayer

voids, and enhanced interlayer adhesion. However, the coefficient of friction increased with extrusion temperature, showing that lower wear does not necessarily correspond to lower friction. This is an important point in polymer tribology, since friction and wear are tribosystem-dependent responses controlled by the interaction among surface topography, counterbody, debris, and transfer film.

The internal architecture of printed PLA also affects its tribological response. Studies on infill density and infill pattern have shown that higher internal support can reduce localized deformation and improve wear resistance, provided that the printed structure promotes effective load distribution [4–6]. Srivastava and Singh [6], using Taguchi/ANOVA optimization, identified printing orientation as the most influential parameter for the tribological performance of PLA, followed by layer thickness, infill pattern, and infill density. Thus, the wear performance of neat PLA should be interpreted as the result of the combined effect of material structure, printing strategy, and sliding conditions.

This interpretation also provides the basis for understanding the role of reinforcements in printed PLA. If the wear response of neat PLA is governed by how the printed architecture supports load, accommodates deformation, and stabilizes debris or tribological layer, the addition of fillers should be viewed as an additional structural variable within this same tribological system. Reinforcements can modify the local stiffness, interfacial strength, thermal dissipation, and third-body behavior of the printed material, thereby changing the way the contact evolves during sliding [7].

The incorporation of reinforcements into PLA has been investigated as a strategy to improve its wear resistance. Particulate, fibrous, and lamellar fillers can increase stiffness, strength, load-bearing capacity, and transfer-film stability [8–15]. However, the tribological

efficiency of a reinforcement depends not only on its presence, but also on its dispersion, morphology, interfacial adhesion with the PLA matrix, and ability to participate in a stable tribological layer. In this sense, reinforced PLA should be understood as a matrix–filler–interface system, in which wear resistance is controlled by the balance between load support, filler retention, debris generation, and third-body behavior.

Graphene is particularly attractive as a PLA reinforcement because of its high aspect ratio, high stiffness, thermal conductivity, and lamellar solid-lubricating character. Liesenfeld et al. [8] showed that graphene incorporation into FFF-printed PLA improves stiffness, strength, hardness, and electrical conductivity, while also modifying the crystallization behavior of the PLA matrix. These previously reported structural and mechanical changes may improve load-bearing capacity and restrict matrix deformation. Their possible tribological implications, including the contribution of graphene to interfacial shearing and protective-layer development, are supported by previous tribological studies on graphene- and carbonaceous-filler-reinforced PLA [7,9,12,25].

Tribological studies on graphene-reinforced PLA indicate that graphene can reduce wear when the platelets are properly distributed and retained within the matrix. Rachaiah et al. [9] reported a significant increase in wear resistance for graphene-reinforced PLA compared with neat PLA, attributing this behavior to the formation of protective layers during sliding and to the lubricating effect of graphene. Avalappa et al. [10] also showed that layer height, printing temperature, and printing speed significantly affect surface roughness and wear loss in graphene-reinforced PLA. Therefore, the beneficial effect of graphene remains strongly dependent on the interaction between material formulation, printing parameters, and sliding conditions.

Other reinforcement systems also support this interpretation. Biocarbon-reinforced PLA showed improved tribological resistance with increasing filler content [11], while comparative

studies on carbonaceous fillers demonstrated that filler morphology governs transfer-film formation and wear mechanisms in additively manufactured PLA [12]. Particulate silicon reinforcement also reduced specific wear rate and coefficient of friction in PLA, mainly due to improved load-bearing capacity and tribofilm formation [13]. In fiber-reinforced PLA systems, wear reduction depends strongly on the fiber–matrix interface, since fiber pull-out and hard debris formation may promote third-body abrasion if interfacial bonding is poor [14,15].

Although reinforcement is an effective route to improve the tribological response of printed PLA, filler addition alone does not fully address the processing-induced limitations of FFF parts. The performance of both neat and reinforced PLA remains strongly affected by the thermal history imposed during deposition, especially the rapid cooling of the extruded roads, which limits crystallization, preserves residual stresses, and may restrict interlayer bonding. Therefore, post-processing treatments become relevant because they can modify the polymer matrix after printing, improving the structural conditions under which the filler or the neat PLA surface participates in the sliding contact.

Annealing is therefore a relevant post-processing strategy for tailoring the structure of FFF-printed PLA-based materials. By promoting molecular rearrangement and crystallization after printing, heat treatment can modify matrix stiffness, strength, dimensional stability, and the degree of structural continuity between deposited roads [16]. From a tribological perspective, these changes may improve the ability of the surface to sustain contact stresses and limit excessive plastic deformation during sliding. However, the same increase in crystallinity and stiffness may also reduce damage tolerance, making the final wear response dependent on the balance between improved load support, matrix embrittlement, filler retention, and transfer-film stability.

For both neat and graphene-reinforced PLA, Liesenfeld et al. [16] demonstrated that annealing increased crystallinity, thermal stability, and mechanical properties. These previously reported structural and mechanical changes provide a basis for interpreting the tribological results obtained in the present study. The possible contribution of graphene to interfacial shearing and tribological-layer formation is supported by previous tribological studies [7,9,12,25].

Nevertheless, the effect of annealing on wear performance should not be considered automatically beneficial. Higher crystallinity and stiffness may be accompanied by reduced ductility and damage tolerance, which can promote microcracking and brittle debris formation during sliding. Therefore, the improvement in wear resistance depends on achieving a balance between increased load-bearing capacity, preserved interlayer integrity, filler retention, and transfer-film stability. In neat PLA, annealing may reduce surface deformation and improve dimensional stability. In graphene-reinforced PLA, annealing may additionally improve matrix–filler interaction and favor the development of an effective tribological layer. However, excessive heat treatment may increase matrix embrittlement, filler pull-out, and abrasive third-body action.

Overall, the literature shows that the wear behavior of neat FFF-printed PLA is strongly controlled by printed architecture and transfer-film stability. It also shows that graphene-reinforced PLA has potential for improved wear resistance due to increased stiffness, higher load-bearing capacity, and the lubricating/protective action of graphene platelets. Annealing appears as a promising strategy to further improve structural organization, crystallinity, strength, and dimensional stability. However, the combined effect of annealing and graphene reinforcement on tribological performance remains insufficiently understood.

Accordingly, this study investigates how annealing temperature and time influence the tribological behavior of FFF-printed neat and graphene-reinforced PLA, using the as-printed

condition as a reference. Particular attention is given to the relationships among annealing parameters, crystallinity, mechanical properties, worn-surface morphology, and the resulting wear mechanisms. By clarifying how thermal post-processing modifies the wear response of both materials, this study aims to support the definition of suitable annealing routes for expanding the application of PLA-based materials in functional components subjected to sliding contact.

2. Materials and methods

2.1. Materials, additive manufacturing and annealing treatment

Commercial PLA-based filaments were used in this study: neat PLA and graphene-reinforced PLA, both with a nominal diameter of 1.75 mm. The specimens were manufactured by fused filament fabrication (FFF) using a 0.4 mm nozzle, an extrusion temperature of 200 °C, a heated bed temperature of 60 °C, a layer thickness of 0.25 mm, a printing speed of 60 mm/s, a raster orientation of 45°/135°, and 100% infill density. These printing parameters were kept constants for all specimens to minimize processing-related variability among the investigated conditions.

After printing, part of the specimens was subjected to annealing heat treatment in an electric furnace. The heat treatments were performed at 90, 100, and 120 °C for 60, 120, and 240 min. After the holding time, the samples were cooled to room temperature inside the furnace to reduce thermal gradients and avoid excessive warping. Untreated specimens were maintained as reference conditions for comparison with the annealed materials.

2.2. Tribological testing

The wear behavior of the printed specimens was evaluated using a reciprocating ball-on-flat configuration, according to ASTM G133. The tests were performed using a 6 mm diameter Al_2O_3 ball as the counterbody. All experiments were conducted under dry sliding conditions at room temperature, approximately 20 °C, and relative humidity close to 50%.

The reciprocating tests were carried out under normal load of 10 N, with a frequency of 2 Hz, a stroke length of 10 mm, and a total sliding distance of 70 m. These parameters corresponded to 3600 reciprocating cycles for each test condition. During the tests, the coefficient of friction was continuously recorded as a function of sliding distance, allowing the evaluation of the running-in stage and the steady-state friction regime.

Wear was quantified based on the mass loss of the specimens. Each sample was weighed before and after testing using a high-precision analytical balance with a resolution of 0.00001 g. Before and after the tribological tests, the specimens were cleaned in an ultrasonic bath to remove loose wear debris and surface contaminants and subsequently dried using a hot-air blower. After drying, the specimens were allowed to equilibrate for 60 s before weighing. Each mass measurement was repeated three times, and the mean value was used to determine the mass loss.

The density of each material and annealing condition was experimentally determined according to Archimedes' principle by measuring the specimen mass in air and its apparent mass while immersed in distilled water. The density was obtained from the specimen mass in air and the difference between the measurements in air and in water, considering the density of water at the test temperature. The worn volume was then calculated by dividing the measured mass loss by the corresponding experimentally determined density. The specific wear coefficient was obtained by normalizing the worn volume by the applied normal load and the total sliding distance. This

procedure allowed the wear responses of the different materials and annealing conditions to be compared independently of the applied load and sliding distance. Three independent tests were performed for each experimental condition, and the results are reported as mean $\pm$ standard deviation.

2.3. Worn surface analysis

After the tribological tests, the wear tracks were analyzed by stereomicroscopy and scanning electron microscopy (SEM) to identify the dominant wear mechanisms. Stereomicroscopy was initially used to obtain a general overview of the wear track morphology, including track continuity, apparent width, debris accumulation, surface damage distribution, and macroscopic differences among the tested conditions.

The worn surfaces were subsequently examined by SEM to investigate the local damage mechanisms in greater detail. Prior to SEM analysis, the specimens were coated with a conductive metallic layer. SEM observations were used to identify features such as abrasive grooves, adhesive transfer, plastic deformation, microcracking, delamination, debris compaction, material pull-out, and formation or disruption of compacted tribological surface layers.

2.4. Data used for the correlation analysis

To support the interpretation of the tribological results, previously published thermal and mechanical data obtained for the same neat PLA and graphene-reinforced PLA materials, printing parameters, and annealing conditions were incorporated into the present correlation analysis [16].

The thermal dataset comprised the degree of crystallinity determined by differential scanning calorimetry. The mechanical dataset included tensile strength, tensile elastic modulus, tensile strain at break, flexural strength, flexural modulus, flexural strain, J-integral, and stress

intensity factor. These properties were selected because they describe the structural organization, stiffness, load-bearing capacity, deformation response, and resistance to crack initiation of the printed materials, which are directly relevant to the mechanisms involved in material removal during reciprocating sliding.

The values used in this work correspond to the mean results obtained from five specimens for each material and annealing condition, with the respective standard deviations. The complete experimental procedures, calculation methods, and detailed discussion of the thermal and mechanical characterization were reported previously in Ref. [16].

In the present study, the specific wear coefficient was calculated by dividing the worn volume by the product of the applied normal load and the total sliding distance. These data were then used exclusively to establish phenomenological relationships between the specific wear coefficient and the degree of crystallinity, tensile and flexural properties, and fracture-mechanics parameters. This approach enabled the tribological response and worn-surface morphology to be interpreted in relation to annealing-induced changes in matrix organization, load support, deformation resistance, and susceptibility to crack formation.

3. Results and Discussion

3.1 Effect of annealing on tribological behavior

Figure 1 shows the combined influence of annealing temperature and time on the specific wear coefficient of neat and graphene-reinforced PLA. Before annealing, the as-printed neat PLA and graphene-reinforced PLA exhibited specific wear coefficients of approximately 5.92×10^{-3} and $2.05 \times 10^{-3} \text{ mm}^3/(\text{N}\cdot\text{m})$, respectively. For neat PLA, the values within the investigated annealing domain ranged from approximately $5.11 \times 10^{-3} \text{ mm}^3/(\text{N}\cdot\text{m})$ under the mildest annealing

condition to $1.56 \times 10^{-3} \text{ mm}^3/(\text{N}\cdot\text{m})$ under the most severe condition. These values correspond to reductions of approximately 14% and 74%, respectively, relative to the as-printed condition. Considering only the annealed conditions, the decrease from 5.11×10^{-3} to $1.56 \times 10^{-3} \text{ mm}^3/(\text{N}\cdot\text{m})$ represents a reduction of approximately 69%. A more pronounced response was observed for graphene-reinforced PLA, for which the specific wear coefficient ranged from approximately 1.35×10^{-3} to $0.14 \times 10^{-3} \text{ mm}^3/(\text{N}\cdot\text{m})$. Relative to the as-printed graphene-reinforced PLA, these values correspond to reductions of approximately 34% and 93%, respectively, while the reduction across the investigated annealing domain was nearly 90%. Overall, annealing improved the wear resistance of both materials throughout the investigated conditions, with the greatest reduction obtained for graphene-reinforced PLA under the most severe treatment.

The structural and mechanical changes previously measured for these materials after annealing [16] provide a basis for interpreting the wear reduction observed in the present study. The current correlations suggest that the resulting increases in crystallinity, strength, and stiffness improved load support and restricted excessive near-surface deformation, thereby reducing material removal during sliding. However, these mechanisms should be considered a literature-supported interpretation and will be more directly assessed through the correlations with crystallinity and mechanical properties presented in the following sections.

Graphene-reinforced PLA exhibited a substantially lower specific wear coefficient than neat PLA throughout the investigated annealing domain. The lower wear of graphene-reinforced PLA is consistent with previous studies on graphene- and carbonaceous fillers-reinforced PLA [7,9,12]. Possible contributing mechanisms include increased load-bearing capacity, restricted matrix deformation, and changes in debris behavior or tribological-layer development. These

mechanisms are discussed here as possible interpretations and should not be considered direct evidence of graphene-mediated transfer-film formation.

Figure 2 and Figure 3 show the evolution of the friction coefficient for the annealed and as-printed materials. In all cases, the friction curves were characterized by an initial running-in stage followed by a comparatively stable friction regime. During running-in, the friction coefficient generally increased with sliding distance until reaching a quasi-steady state. The as-printed specimens exhibited the same overall behavior, indicating that annealing modified the friction level rather than the fundamental friction evolution. This initial increase can be associated with the progressive accommodation of the contacting asperities, enlargement of the real contact area, modification of the original printed surface topography, and the generation and redistribution of wear debris. As sliding proceeds, the interaction among the polymer surface, the Al_2O_3 counterbody, and the evolving third-body material gradually establishes a relatively stable friction regime.

The neat PLA curves in Figure 2 exhibited a stronger dependence on annealing conditions than those of graphene-reinforced PLA. The as-printed material reached a steady-state friction coefficient of approximately 0.59, which was intermediate between the values measured for the annealed conditions. Annealing at 90 °C generally reduced the friction coefficient, yielding steady-state values between approximately 0.51 and 0.55, whereas treatments at 100 and 120 °C progressively increased the friction coefficient, reaching values of approximately 0.65–0.73 under the most severe conditions. Increasing the annealing time also tended to increase friction, particularly at the higher annealing temperatures.

The graphene-reinforced PLA exhibited a similar friction evolution, although within a considerably narrower range (Figure 3). The as-printed composite presented a steady-state friction

coefficient close to 0.49, very similar to the values obtained after annealing. Regardless of the thermal treatment, the friction coefficient remained approximately between 0.48 and 0.54, and the influence of annealing time was comparatively small, especially at 120 °C, where the three curves converged toward similar steady-state values. These results show that graphene-reinforced PLA exhibited not only a lower specific wear coefficient but also a lower sensitivity of friction to the investigated annealing parameters than neat PLA.

The recurrent rises and sharp decreases observed in Figure 2 and Figure 3 are associated with the reciprocating nature of the test. At each end of the stroke, the sliding velocity decreases to zero and subsequently changes direction. At the same time, the tangential force reverses its sign. Consequently, the friction signal passes through a transient region involving pre-sliding, near-static contact, and renewed dynamic sliding. When the coefficient is presented as a positive magnitude, the tangential-force reversal may appear as a narrow decrease in the recorded friction coefficient. Reciprocating contacts are therefore intrinsically affected by repeated transitions between near-zero velocity and dynamic sliding [17,18].

The broader cyclic increases and decreases superimposed on these reversal transients may also be related to the evolution of wear debris and tribological layer. Wear particles can accumulate and compact within the contact, temporarily increasing the interfacial shear resistance. Subsequent fragmentation, displacement, or ejection of this third-body material changes the real contact conditions and may produce an abrupt decrease in friction. The subsequent generation and compaction of new debris can then cause the friction coefficient to increase again. Previous investigations on polymer composites have demonstrated that friction and wear evolve as transient processes controlled by the accumulation, consolidation, adherence, disruption, and renewal of tribological layer [19,20].

Accordingly, the narrow periodic transients are consistent with the reversal of the reciprocating motion associated changes in sliding velocity and tangential-force direction. The broader fluctuations may additionally be influenced by the accumulation, compaction fragmentation, and displacement of wear debris-within the contact. However, the specific contribution of third-body evolution to the friction signal cannot be isolated from the present measurements. The relative contribution of debris accumulation and transfer-film formation to these fluctuations is further examined through the worn-surface and wear-debris analyses presented in Section 3.4.

Figure 4 summarizes the combined effects of annealing temperature and time on the mean friction coefficient calculated over the final 10 m of sliding. For comparison, the as-printed neat PLA exhibited a mean friction coefficient of approximately 0.59, whereas the as-printed graphene-reinforced PLA presented a value close to 0.49. Relative to these reference conditions, annealing affected the two materials differently. For neat PLA, the friction coefficient initially decreased under the mildest annealing conditions, reaching values close to 0.51, but subsequently increased with increasing annealing temperature and time, attaining approximately 0.73 under the most severe treatment. In contrast, graphene-reinforced PLA exhibited only minor variations around the as-printed value, remaining within the relatively narrow interval of 0.48–0.54 throughout the investigated annealing domain. The contour orientation indicates that annealing temperature exerted a stronger influence than annealing time, particularly for neat PLA. These observations are fully consistent with the friction evolutions presented in Figure 2 and Figure 3, where the changes produced by annealing are reflected primarily in the steady-state friction level rather than in the overall shape of the friction curves.

The simultaneous reduction in the specific wear coefficient and increase in the friction coefficient is not contradictory, because friction and wear are distinct responses of the tribological system. Wear is primarily related to the amount of material removed, whereas friction depends on the shear resistance of the instantaneous interface, the real contact area, surface topography, adhesion, and the presence and stability of third-body material. Zainal et al. [3] similarly observed that processing conditions capable of reducing the specific mass loss of FFF-printed PLA could increase its coefficient of friction. Frunzaverde et al. [21] also reported that changes in PLA mesostructure and surface finishing modified friction independently of mechanical strength and material removal. A denser and mechanically stronger structure may therefore resist material removal while still requiring a higher tangential force to sustain sliding.

For graphene-reinforced PLA, the smaller increase in friction and the markedly lower wear coefficient are consistent with the combined reinforcing and solid-lubricating roles attributed to graphene [7,9,12]. The lamellar morphology of graphene can facilitate interfacial shearing, while graphene-containing debris may contribute to a more effective tribological layer. The relatively narrow friction range in Figure 4b suggests that graphene reduced the sensitivity of the contact to annealing time. However, the existence, continuity, composition, and stability of such a layer must be demonstrated by microscopy before a definitive mechanism is assigned.

Overall, the results reveal a clear distinction between the effects of annealing on friction and wear. More severe annealing conditions substantially improved wear resistance, especially for graphene-reinforced PLA, but generally increased the friction coefficient. Therefore, the condition providing the lowest wear coefficient does not necessarily provide the lowest friction coefficient. Selection of the most appropriate annealing condition should consequently consider the relative

importance of wear resistance, frictional resistance, dimensional stability, and damage tolerance for the intended application.

3.2 Influence of crystallinity on tribological performance

Figure 5 shows an inverse relationship between the degree of crystallinity and the specific wear coefficient for both neat and graphene-reinforced PLA. The linear regressions summarized in Table 1, indicate that increasing X_c was associated with a progressive reduction in material loss under dry reciprocating sliding. For neat PLA, the regression provided a good fit to the experimental data ($R^2 = 0.92$), showing that crystallinity accounted for most of the observed variation in the specific wear coefficient within the investigated conditions. Graphene-reinforced PLA exhibited the same decreasing trend, but the lower coefficient of determination ($R^2 = 0.68$) indicates greater data dispersion and suggests that factors other than crystallinity, including graphene dispersion, matrix–filler interfacial interactions, and local structural heterogeneity, also influenced the wear response.

For neat PLA, the equation indicates a reduction of approximately $0.11 \times 10^{-3} \text{ mm}^3/(\text{N} \cdot \text{m})$ in the specific wear coefficient for each percentage-point increase in crystallinity. This dependence is consistent with the results shown in Figure 1, in which more severe annealing conditions produced lower wear. The correlation in Figure 5 indicates that the variation in wear is associated with the structural changes accompanying annealing rather than with annealing temperature or holding time considered solely as processing parameters. Increased crystallinity can enhance matrix stiffness, dimensional stability, and resistance to localized deformation, thereby improving the ability of the printed structure to support the contact load and reducing the volume of material removed during sliding. Ref. [16] previously reported the annealing-induced increases in crystallinity and mechanical performance used in the present correlation. The association of these

changes with reduced material removal is established here from the relationship shown in Figure 5.

The graphene-reinforced PLA data were described by the equation $k = (1.57884 - 0.02055X_c) \times 10^{-3}$, indicating that each percentage-point increase in crystallinity was associated with a reduction of approximately $0.02055 \times 10^{-3} \text{mm}^3/(\text{N}\cdot\text{m})$ in the specific wear coefficient. Despite this lower sensitivity to crystallinity, the composite maintained substantially lower wear coefficients than neat PLA throughout the investigated X_c range.

This result suggests that factors associated with graphene reinforcement contribute to the lower wear beyond the structural changes associated with annealing. Graphene incorporation has been reported to modify PLA crystallization while increasing stiffness, strength, and load-bearing capacity [8,16]. During sliding, graphene platelets and graphene-containing debris may also restrict matrix deformation and contribute to the development of a more protective tribological layer, as previously proposed for graphene-reinforced and carbonaceous-filler-containing PLA systems [7,9,12]. These effects help explain why the composite maintains lower wear even before reaching its highest crystallinity levels.

The lower R^2 obtained for graphene-reinforced PLA further suggests that its wear behavior depends on variables not represented directly by X_c , including platelet dispersion, matrix–graphene adhesion, filler retention, debris evolution, and the formation and stability of the tribological layer. Similarly, the error bars and the dispersion of some experimental points show that specimens with close crystallinity values may exhibit different wear responses. Consequently, crystallinity should be regarded as an important structural descriptor rather than an isolated governing parameter. The final tribological response results from its interaction with the FFF-

generated architecture, mechanical properties, graphene reinforcement, and the transient conditions established at the sliding interface.

Overall, Figure 5 and Table 1 show a clear inverse association between crystallinity and the specific wear coefficient within the investigated annealing conditions. For neat PLA, this correlation is comparatively stronger, whereas graphene-reinforced PLA combines the beneficial effect of matrix crystallization with the reinforcing and potentially tribological action of graphene. This combined contribution explains the markedly lower wear coefficients obtained for the composite throughout the investigated annealing conditions.

3.3 Correlation between mechanical properties and tribological performance

Building on the structural relationship established in Figure 5, Figure 6 to Figure 8 examine whether the annealing-induced changes in mechanical behavior can be used to describe the corresponding variation in wear resistance. Because crystallinity and the mechanical properties changed simultaneously as a consequence of annealing, these regressions should be interpreted as phenomenological correlations rather than as independent cause-and-effect relationships. Nevertheless, the relative strength of the correlations provides useful information on which mechanical responses are more closely associated with material removal under the present sliding conditions.

Figure 6 correlates the specific wear coefficient with the tensile properties of both materials. Clear inverse relationships were obtained for tensile strength and tensile elastic modulus. As summarized in Table 2, tensile strength presented coefficients of determination of 0.907 for neat PLA and 0.839 for graphene-reinforced PLA, while the corresponding values for tensile elastic modulus were 0.833 and 0.799. Thus, the conditions exhibiting greater tensile strength and stiffness consistently showed lower specific wear coefficients. These trends agree

with the mechanical characterization of the same materials, in which annealing increased tensile strength and elastic modulus, particularly at higher temperatures and longer holding times [16].

The correlations suggest that wear resistance is more closely associated with the ability of the printed structure to sustain stress and restrict deformation. A stronger and stiffer matrix is less susceptible to excessive viscoelastic deformation beneath the counterbody and can distribute the contact load more effectively through the deposited roads and their interfaces. Such changes could reduce localized deformation and susceptibility to progressive material detachment under the investigated sliding conditions. The comparatively steep regression slopes obtained for neat PLA also indicate that its wear response is more sensitive to changes in the mechanical condition of the matrix. For graphene-reinforced PLA, the smaller slopes partly reflect its substantially lower initial wear level and the narrower range over which its wear coefficient varied.

In contrast, tensile strain at break exhibited only moderate-to-weak relationships with the specific wear coefficient, with R^2 values of 0.523 for neat PLA and 0.308 for graphene-reinforced PLA. Annealing was previously shown to increase strength and stiffness while slightly reducing tensile strain [16]. Therefore, the apparent association between lower strain at break and lower wear should not be interpreted as evidence that reduced ductility is intrinsically beneficial to tribological performance. Instead, both responses result from the same structural evolution of the material. Tensile strain measured at macroscopic fracture does not directly describe the capacity of the near-surface region to support repeated contact loading, explaining its lower predictive ability.

The flexural correlations in Figure 7 reinforce this interpretation. Flexural strength exhibited the strongest overall relationships with wear, with R^2 values of 0.928 for neat PLA and 0.814 for graphene-reinforced PLA, as shown in Table 3. Flexural modulus also showed

meaningful inverse correlations, with R^2 values of 0.734 and 0.793, respectively. The consistency between the tensile and flexural results demonstrates that wear reduction was not associated with a single loading mode. Rather, it accompanied a general improvement in the load-bearing capacity and rigidity of the printed structure.

Flexural strength may be particularly relevant because bending involves tensile and compressive stresses distributed through the specimen thickness. Although the stress state differs from that established during reciprocating contact, the flexural response is sensitive to local defects, deposited-road cohesion, and interlayer integrity. These same structural features affect the ability of the near-surface region to withstand the combined normal and tangential stresses imposed during sliding. The strong correlation between flexural strength and wear suggests that the reduction in material removal was accompanied by improvements in matrix strength and mechanical continuity of the FFF-generated structure. Liesenfeld et al. [16] reported that annealing increased flexural strength by up to 38.4% for neat PLA and 25.2% for graphene-reinforced PLA, while the flexural modulus increased by up to 22.9% and 14.0%, respectively.

Flexural strain, however, was not a reliable descriptor of the wear response. The regression for neat PLA presented an R^2 value of only 0.001, indicating the absence of a meaningful linear relationship, whereas graphene-reinforced PLA showed a moderate value of 0.518. Consequently, the sign and magnitude of the flexural-strain regression coefficients should not be assigned a direct mechanistic meaning. The results indicate that the capacity to support load before failure, represented by strength and modulus, is more closely associated with wear resistance than the displacement or strain attained at the end of the flexural test.

Figure 8 extends the analysis to the fracture-mechanics parameters obtained from the SENB tests. The specific wear coefficient decreased as both the J -integral and the stress intensity

factor increased. According to Table 4, the relationships with the J -integral were moderate, with R^2 values of 0.584 for neat PLA and 0.493 for graphene-reinforced PLA. Higher coefficients of determination were obtained for the stress intensity factor, reaching 0.753 and 0.687, respectively. These results suggest that resistance to crack initiation and propagation contributes to wear control, although it is not as directly associated with wear as tensile or flexural strength.

During reciprocating sliding, repeated normal and tangential loading can promote the nucleation and growth of subsurface cracks at pores, road boundaries, interlayer interfaces, or other local discontinuities. Higher fracture resistance could reduce the susceptibility to crack coalescence and the detachment of larger fragments. However, subsurface crack initiation and propagation were not directly characterized in the present study. The increase in J and K produced by annealing was attributed to improved contact between deposited layers and to the structural reorganization of the PLA matrix [16]. The inverse relationships in Figure 8 therefore support the interpretation that improved interlayer integrity and crack resistance contribute to the lower material loss observed after annealing.

The stronger correlation obtained for K than for J should nevertheless be interpreted cautiously. In the mechanical characterization, K was calculated from the J -integral and the tensile elastic modulus [16]. It therefore incorporates both the energy associated with crack initiation and the elastic response of the material. Its comparatively higher R^2 may consequently reflect the combined influence of fracture resistance and stiffness rather than an entirely independent relationship with wear.

A comparison of all regressions reveals a consistent hierarchy. Tensile and flexural strength showed the strongest relationships with the specific wear coefficient, followed by the tensile and flexural moduli and the stress intensity factor. The J -integral presented moderate correlations,

whereas tensile and flexural strains were comparatively weak descriptors. Together with Figure 5, these results indicate that the wear improvement produced by annealing is associated with the combined structural and mechanical changes induced by annealing, including increased crystallinity, strength and stiffness. Deformation at macroscopic failure appears to have a secondary role under the investigated tribological conditions.

The distinction between neat and graphene-reinforced PLA is also important. The composite retained substantially lower wear coefficients across the entire range of tensile, flexural, and fracture-mechanics properties. In several regions of Figure 6 to Figure 8, the mechanical-property ranges of the two materials approach or overlap, while their wear levels remain clearly separated. Therefore, the superior wear resistance of graphene-reinforced PLA cannot be explained solely by differences in bulk mechanical properties. The persistent difference in wear between the two materials suggests contributions not captured by the bulk mechanical properties considered in the present correlations. Based on previous studies, these contributions may involve local reinforcement, differences in matrix deformation and debris behavior, and the possible development of a more protective tribological layer [7,9,12]. This also explains why the composite generally exhibited smaller regression slopes: its wear response was less dependent on changes in a single matrix property because additional filler-related mechanisms were active.

Overall, Figure 5 to Figure 8 and Table 2 to Table 4 show that wear evolution was strongly associated with the coupled structural and mechanical changes induced by annealing. Increased crystallinity was accompanied by improved strength, stiffness, interlayer continuity, and resistance to crack initiation, collectively reducing the susceptibility to material removal. For neat PLA, the comparatively strong crystallinity and strength correlations suggest that matrix reorganization was more closely associated with the wear response. For graphene-reinforced PLA, the same

annealing-induced improvements were superimposed on the reinforcing and tribological effects of graphene. The relative contributions of cracking, debris retention, material pull-out, and transfer-layer development are examined through the worn-surface observations in the following section.

3.4 Worn-surface morphology and wear mechanisms

The stereomicrographs in Figure 9 provide a macroscopic representation of the changes in wear-track dimensions produced by annealing and graphene incorporation. For the detailed SEM analysis presented in Figure 10, three representative annealing conditions were selected to span the range of wear severity observed experimentally: 120 °C for 240 min, corresponding to the least severe wear condition; 100 °C for 120 min, representing an intermediate condition; and 90 °C for 60 min, corresponding to the most severe wear condition. This selection enabled the evolution of the dominant wear mechanisms with increasing specific wear coefficient to be examined. Under all annealing conditions, graphene-reinforced PLA exhibited substantially narrower wear tracks than neat PLA.

The macroscopic morphology also reveals differences in the spatial distribution of damage. Neat PLA generally displayed broad and diffuse wear tracks, with irregular boundaries and extensive regions in which the original FFF surface texture was flattened or obscured. These features indicate that the contact affected several adjacent deposited roads and promoted substantial lateral displacement of the polymer. Conversely, the graphene-reinforced specimens exhibited more sharply defined and continuous tracks, generally confined to a narrower region. Local bright deposits along or within some of the dark tracks indicate the presence of accumulated or compacted debris. It should be emphasized that track width alone does not represent the removed volume, because tracks of similar width may differ in depth and in the amount of material

displaced toward their edges. Nevertheless, the morphological trends are consistent with the substantially lower specific wear coefficients measured for the composite.

The SEM observations in Figure 10 demonstrate that material removal did not occur through a single mechanism. Instead, the worn surfaces contain evidence of combined abrasive action, adhesive deformation, surface fatigue, microcracking, delamination, debris compaction, and localized material pull-out. In polymeric contacts, the relative contribution of these mechanisms depends on the penetration of counterbody asperities, the ability of the surface to deform, the cohesive resistance of the polymer, and the behavior of detached material within the contact. Parallel grooves are typically associated with microploughing or microcutting, whereas smooth smeared regions and shear-aligned ridges indicate plastic flow. Plate-like fragments, lifted surface layers, and cavities are characteristic of delamination or adhesive tearing, while narrow cracks and interconnected crack networks indicate cyclic surface or subsurface fatigue. Compacted patches formed by fine particles are evidence of the participation of wear debris as a third body [22–24].

For neat PLA annealed at 90 °C for 60 min, Figure 10(a1) shows a broad wear track containing extensive flattened regions, irregular surface patches, and large plate-like fragments. At higher magnifications, the surface presents pronounced directional deformation, folded or raised lips, shallow parallel scratches, and locally torn layers. The flattened and wavy regions indicate viscoelastic–plastic flow of the PLA under the combined normal and tangential stresses. The raised lips and smeared patches suggest the formation and subsequent rupture of adhesive junctions between the polymer surface and the counterbody. Repeated junction formation and fracture can stretch the softened near-surface material, producing the observed shear-aligned morphology and eventually generating detached fragments [23,24].

The comparatively large flakes and the cavities beneath partially detached layers in Figure 10(a1) indicate that adhesive deformation was accompanied by delamination. Under reciprocating loading, cracks can initiate at near-surface discontinuities, deposited-road boundaries, residual pores, or interfaces between differently oriented raster roads. Their progressive propagation parallel to the surface isolates thin layers that are subsequently lifted and detached. The sharp boundaries of some fragments and the presence of cracks beneath apparently continuous surface regions support this fatigue-assisted delamination mechanism. Fine longitudinal scratches superimposed on the plastically deformed surface indicate simultaneous microabrasion by asperities of the harder Al_2O_3 counterbody and by detached particles entrapped in the contact.

The graphene-reinforced PLA annealed under the same condition, Figure 10(a2), exhibits a markedly narrower track and a more continuous worn surface. The wavy and shear-aligned texture visible at 100–1000 $\times$ indicates deformation of a thin near-surface layer, but the deformation is more uniformly distributed than in neat PLA. Large-scale removal is restricted to isolated regions, including a local plate-like fragment and a pronounced crack. Other areas show fine debris pressed against the surface and a relatively continuous textured layer. This morphology is consistent with the formation of a compacted third-body layer that partially separates the polymer matrix from direct interaction with the counterbody.

Graphene can contribute to this response through two complementary effects. Its reinforcing action restricts penetration and large-scale deformation of the PLA matrix, whereas the low shear resistance associated with its lamellar structure may facilitate sliding within graphene-containing debris or surface layers [7,9,12,25]. The observed morphology does not permit individual graphene platelets to be identified directly; therefore, their participation should be inferred from the collective response of the composite rather than from isolated features in the

micrographs. The local cracks and detached flake in Figure 10(a2) also demonstrate that graphene reduced, but did not completely suppress, fatigue and delamination under the mildest annealing condition.

For neat PLA annealed at 100 °C for 120 min, Figure 10(b1) shows extensive roughening, overlapping lamellar fragments, raised edges, cavities, and irregular debris distributed across the track. Several fragments appear partially attached to the surface, with their edges bent or lifted in the direction of shear. These features indicate adhesive tearing followed by the progressive delamination of the deformed surface layer. The cavities and dark recesses adjacent to the flakes are consistent with material pull-out after the coalescence of near-surface cracks. Fine grooves running through the underlying surface indicate that abrasion continued after the removal of the upper layer.

Compared with the 90 °C–60 min condition, the intermediate annealing treatment increased the stiffness and strength of the matrix, limiting extensive bulk deformation. However, the lower capacity for deformation associated with the more organized PLA structure can concentrate stresses at local defects and deposited-road interfaces. Consequently, damage becomes increasingly localized in cracks, lifted lamellae, and plate-like debris rather than being accommodated only by broad plastic smearing. This explains why surface features associated with delamination may remain evident even though the gravimetric wear coefficient decreases: the damage becomes more spatially confined, and the total volume detached per sliding cycle is reduced.

The corresponding graphene-reinforced specimen in Figure 10(b2) presents a substantially smoother and more homogeneous track. Most of the worn region is covered by fine wavy ridges and shallow scratches, while severe damage is restricted to isolated pits, short cracks, and small

debris agglomerates. The absence of extensive lifted layers indicates that large-scale delamination was inhibited. The predominant micromechanisms were therefore mild microploughing, limited surface fatigue, and localized third-body abrasion. The isolated pits may correspond to the pull-out of small matrix fragments or compacted debris rather than to the direct pull-out of individual graphene platelets, which cannot be resolved at the magnifications employed.

The intermediate graphene-reinforced condition also shows local patches with a smoother appearance than the surrounding surface. Together with the compacted debris and the continuity of the shear-oriented texture, these regions suggest that detached particles were repeatedly comminuted, redistributed, and pressed against the track. Such a layer can act simultaneously as a protective third body and as a source of frictional resistance. Its formation limits direct penetration by the counterbody and reduces material removal, while its shear and repeated renewal contribute to the measured friction force. This interpretation is consistent with the transient formation, compaction, fracture, and redistribution of polymeric tribological layer described for dry sliding contacts [19,20,23].

The most severe annealing condition produced a distinct evolution in the morphology of neat PLA. In Figure 10(c1), the track is considerably narrower than in Figure 10(a1) and (b1), confirming the improvement in load-bearing capacity inferred from the crystallinity and mechanical-property correlations. Nevertheless, the SEM images show pronounced longitudinal grooves, shear-aligned ridges, local spallation, cracks, and fragmented material near the track edges. The continuous parallel grooves are characteristic of microploughing and localized microcutting. The harder and more dimensionally stable annealed matrix limited deep penetration and broad lateral flow, but the reduction in deformation capacity favored localized fracture when the accumulated strain exceeded the cohesive resistance of the near-surface region.

The lifted layers and sharp-edged cavities in Figure 10(c1) indicate that surface fatigue and delamination remained active. The repeated reversal of the sliding direction subjects previously deformed ridges and surface layers to alternating shear. This cyclic loading promotes crack initiation at the base of the ridges and along weak interfaces, followed by progressive crack growth and eventual spallation. Thus, increasing annealing severity changed the balance among the mechanisms: broad plastic deformation and adhesive smearing became less dominant, whereas localized abrasion, microcracking, and fatigue-assisted delamination became proportionally more evident. This transition is consistent with the classical observation that polymer wear can shift between asperity-scale cutting, ploughing, tearing, and surface fatigue depending on counterface topography and the mechanical response of the polymer [22].

Graphene-reinforced PLA annealed at 120 °C for 240 min, Figure 10(c2), exhibited the most homogeneous worn surface among the conditions examined. The track contains predominantly fine ripple-like textures and shallow shear-aligned grooves, with only isolated adhered fragments, short cracks, and local edge damage. The limited depth and continuity of the grooves indicate mild microploughing rather than extensive microcutting. The finely textured and relatively continuous surface suggests that a thin layer of plastically accommodated and compacted material remained within the contact, protecting the underlying printed structure.

Although the composite surface was comparatively stable, the cracks surrounding some compacted patches show that severe annealing did not eliminate brittle or fatigue-related damage. A compacted third-body layer can carry part of the contact load, but differences in stiffness and deformation between this layer and the underlying matrix can generate interfacial stresses. Local fracture of the layer exposes the matrix, after which new debris is generated and incorporated into a renewed surface layer. This cyclic disruption and reformation provides a morphological basis for

the broader friction fluctuations superimposed on the stroke-reversal transients discussed for Figure 2 and Figure 3.

The morphology of Figure 9 and Figure 10 also helps explain why annealing simultaneously reduced wear and increased friction. The annealed specimens exhibited surface morphologies consistent with reduced penetration, plastic displacement, and material detachment compared with the less severely treated conditions. At the same time, the enlarged effective contact area of the flattened surface and the shear resistance of compacted debris or tribological layers could increase the tangential force required to sustain sliding. Therefore, a surface may exhibit higher friction while losing less material. The local agglomerates, adhered patches, smeared regions, and fractured surface layers observed by SEM are consistent with a continuously evolving third body that alternately adheres, compacts, fractures, and is displaced from the contact [19,20,23,24].

Overall, neat PLA was characterized by a mixed adhesive–abrasive wear regime with extensive plastic flow, surface tearing, fatigue cracking, and delamination. Increasing annealing severity reduced the width of the affected region and suppressed broad viscoelastic deformation, but promoted a relative transition toward localized microabrasion and brittle fatigue damage. Graphene reinforcement changed this balance by restricting matrix deformation, reducing the penetration of the counterbody, limiting the formation of large delaminated fragments, and favoring finer debris and a more continuous tribological surface layer. Consequently, graphene-reinforced PLA exhibited predominantly mild microploughing, localized cracking, and limited delamination, in agreement with its substantially lower specific wear coefficient.

The presence of compacted surface layers is supported by the SEM morphology; however, a definitive identification of a transfer film on the Al_2O_3 counterbody would require direct

examination of the ball by SEM combined with compositional or spectroscopic analysis. Therefore, the features observed in Figure 10 should be described as compacted debris or a tribological layer on the polymer track, while transfer to the counterbody remains a literature-supported mechanism rather than a directly demonstrated result.

4. Conclusions

The influence of annealing on the dry reciprocating tribological behavior of FFF-printed neat PLA and graphene-reinforced PLA was systematically investigated by correlating tribological performance with crystallinity, mechanical properties, and worn-surface morphology. Based on the experimental results, the following conclusions can be drawn:

- Annealing improved the wear resistance of both materials. Relative to the as-printed condition, the specific wear coefficient was reduced by up to approximately 74% for neat PLA and 93% for graphene-reinforced PLA. Considering only the annealed conditions, the reductions across the investigated annealing domain were approximately 69% and 90%, respectively.
- The coefficient of friction increased with annealing severity, by approximately 43% for neat PLA and 11% for graphene-reinforced PLA, showing that the conditions producing lower wear did not necessarily produce lower friction.
- The reduction in wear was associated with annealing-induced changes in crystallinity and mechanical properties. Among the evaluated properties, tensile and flexural strengths exhibited the strongest inverse correlations with the specific wear coefficient.

- Graphene-reinforced PLA exhibited lower wear than neat PLA under all evaluated conditions, even at similar crystallinity and mechanical-property levels, indicating an additional contribution from graphene to the tribological response.
- Neat PLA showed mixed adhesive–abrasive wear, with plastic deformation, delamination, microcracking, and material pull-out. Graphene reinforcement reduced severe surface damage and promoted predominantly mild microploughing, localized cracking, and compacted debris layers.
- Under the investigated test conditions, annealing and graphene reinforcement were associated with substantial improvements in the dry reciprocating sliding wear resistance of FFF-printed PLA.

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Author Contributions

Samara Herrmann: Investigation, Formal analysis, Data curation. Carlos Eduardo Fracari do Nascimento: Investigation, Formal analysis, Data curation. Bruna Karine dos Santos: Investigation, Formal analysis, Data curation. Rafael Luciano Dalcin: Investigation, Formal analysis, Data curation, Writing – review & editing. Alexandre Aparecido Buenos: Validation, Investigation support, Writing – review & editing. Cristiano José Scheuer: Conceptualization, Methodology, Supervision, Project administration, Funding acquisition, Writing – original draft. All authors contributed to the interpretation of the results, reviewed the manuscript, approved the final version, and agreed to be accountable for the content of the work.

Declaration of AI-assisted technologies in the manuscript preparation process

During the preparation of this work, the authors used OpenAI's ChatGPT to assist with language refinement, improvement of clarity and readability. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

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Lista of tables

Table 1. Linear equations describing the variation of the specific wear coefficient as a function of the degree of crystallinity for neat and graphene-reinforced PLA.

Material condition	Linear regression equation	(R ²)
Neat PLA	$k = (8.43 - 0.11X_c) \times 10^{-3}$	0.92
Graphene-reinforced PLA	$k = (1.57 - 0.020X_c) \times 10^{-3}$	0.688

Table 2. Linear equations describing the variation of the specific wear coefficient as a function of the tensile properties for neat and graphene-reinforced PLA.

Tensile property	Material condition	Linear regression equation	R ²
Tensile strength, (σ_t)	Neat PLA	$k = (12.53 - 0.12\sigma_t) \times 10^{-3}$	0.90
	Graphene-reinforced PLA	$k = (3.10 - 0.03\sigma_t) \times 10^{-3}$	0.83
Tensile elastic modulus, (E_t)	Neat PLA	$k = (14.67 - 4.61E_t) \times 10^{-3}$	0.83
	Graphene-reinforced PLA	$k = (3.19 - 0.69E_t) \times 10^{-3}$	0.79
Tensile strain, (ε_t)	Neat PLA	$k = (-1.34 + 2.48\varepsilon_t) \times 10^{-3}$	0.52
	Graphene-reinforced PLA	$k = (-3.15 + 1.80\varepsilon_t) \times 10^{-3}$	0.30

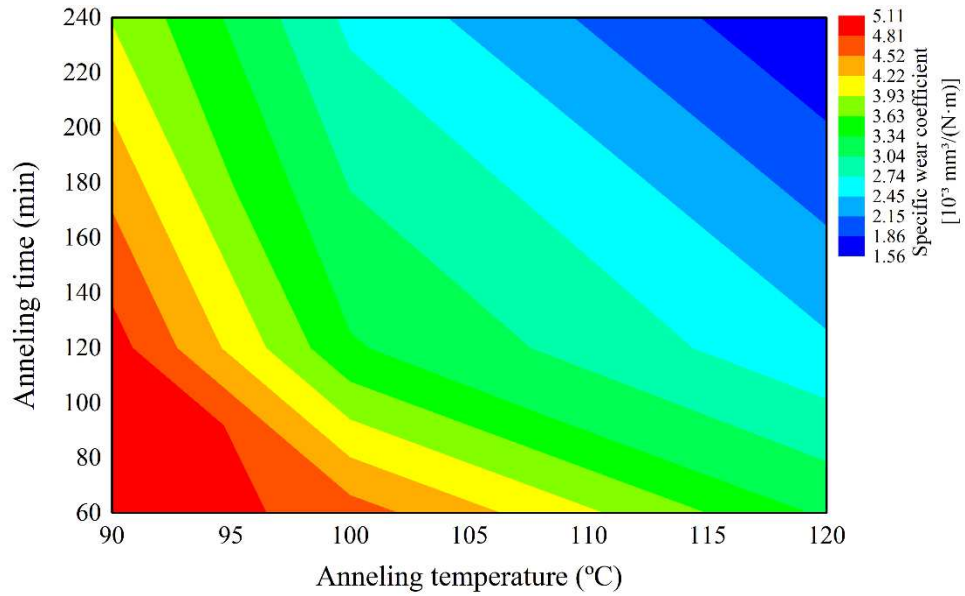
Table 3. Linear equations describing the variation of the specific wear coefficient as a function of the flexural properties for neat and graphene-reinforced PLA.

Flexural property	Material condition	Linear regression equation	R ²
Flexural strength, (σ_f)	Neat PLA	$k = (18.21 - 0.16\sigma_f) \times 10^{-3}$	0.928
	Graphene-reinforced PLA	$k = (5.27 - 0.05\sigma_f) \times 10^{-3}$	0.814
Flexural modulus, (E_f)	Neat PLA	$k = (23.78 - 7.39E_f) \times 10^{-3}$	0.734
	Graphene-reinforced PLA	$k = (8.04 - 2.19E_f) \times 10^{-3}$	0.793
Flexural strain, (ε_f)	Neat PLA	$k = (5.55 - 0.82\varepsilon_f) \times 10^{-3}$	0.001
	Graphene-reinforced PLA	$k = (2.65 - 0.63\varepsilon_f) \times 10^{-3}$	0.518

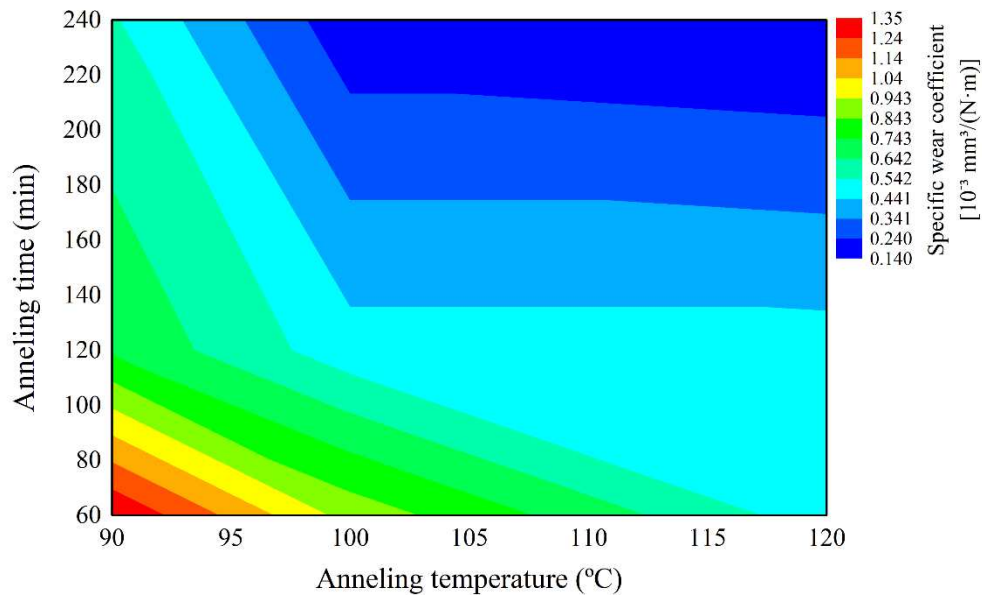
Table 4. Linear equations describing the variation of the specific wear coefficient as a function of the fracture-mechanics parameters for neat and graphene-reinforced PLA.

Fracture-mechanics parameter	Material condition	Linear regression equation	R ²
J-integral, (J)	Neat PLA	$k = (7.52 - 0.29J) \times 10^{-3}$	0.584
	Graphene-reinforced PLA	$k = (1.44 - 0.08J) \times 10^{-3}$	0.493
Stress intensity factor, (K)	Neat PLA	$k = (7.58 - 0.22K) \times 10^{-3}$	0.753
	Graphene-reinforced PLA	$k = (1.48 - 0.04K) \times 10^{-3}$	0.687

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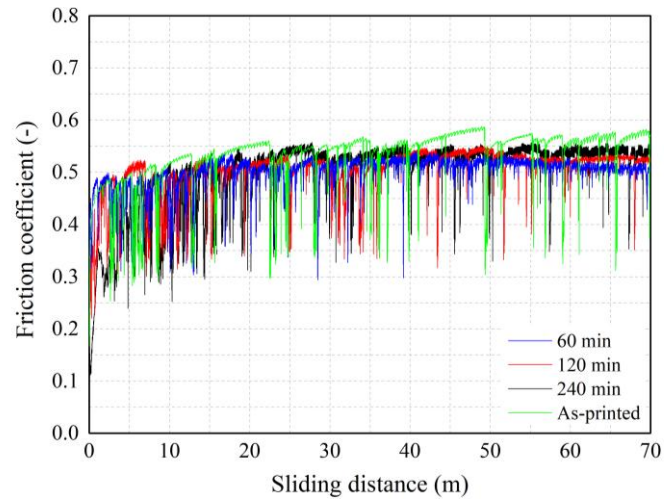


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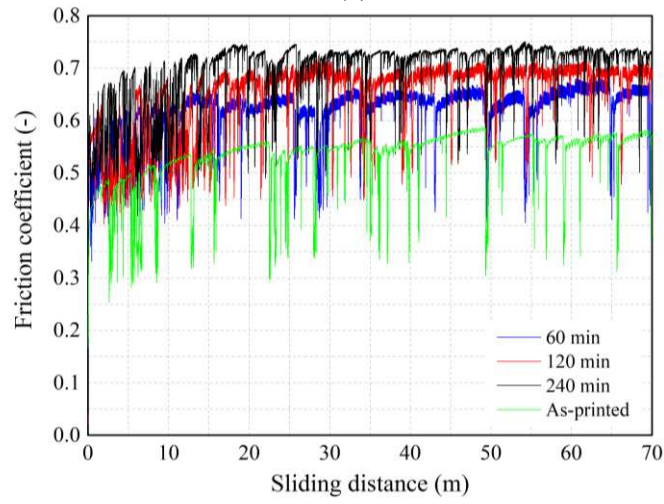


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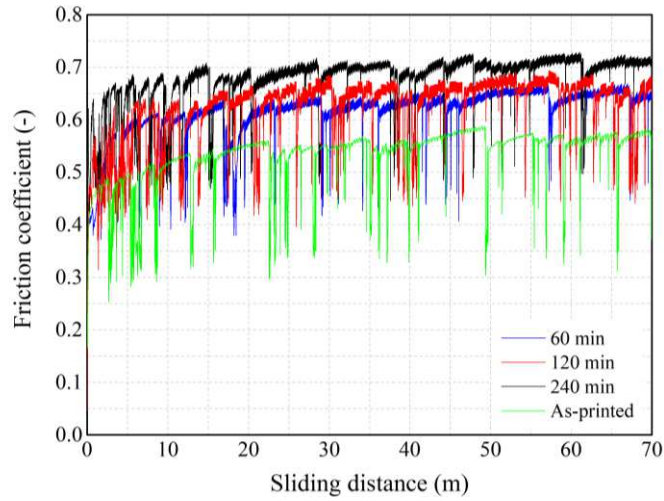
Figure 1. Effect of annealing temperature and time on the specific wear coefficient of FFF-printed (a) neat PLA and (b) graphene-reinforced PLA.



(a)

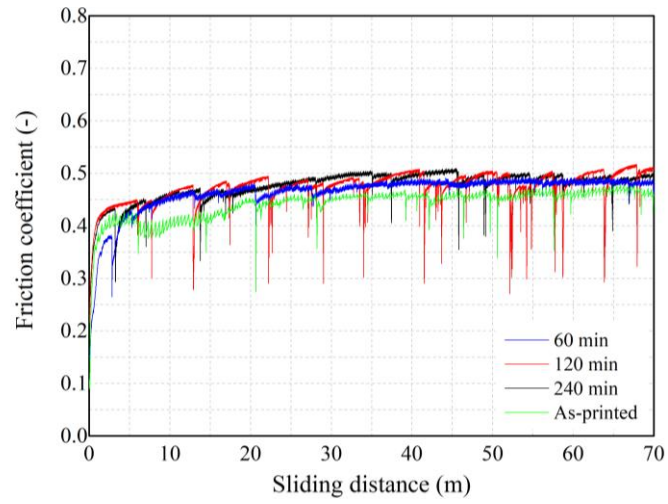


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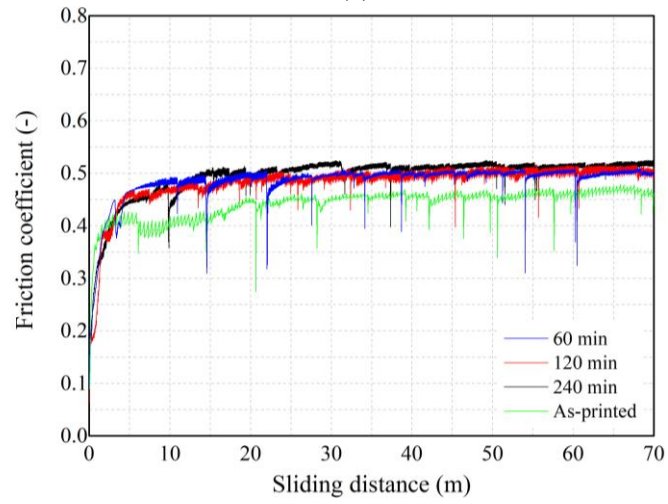


(c)

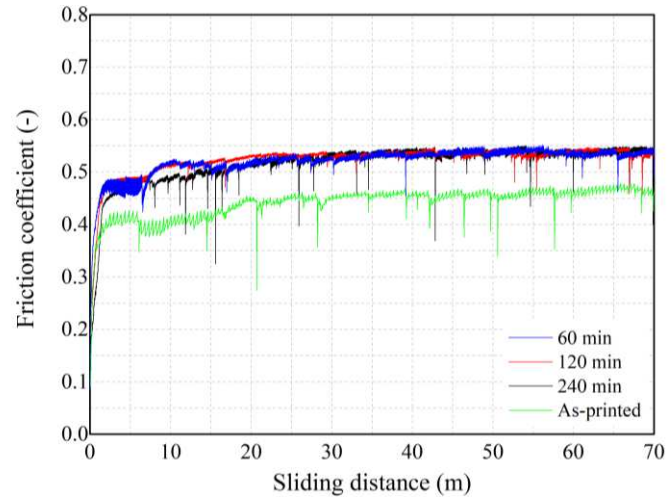
Figure 2. Evolution of the friction coefficient as a function of sliding distance for neat PLA specimens annealed for 60, 120, and 240 min at: (a) 90 °C, (b) 100 °C, and (c) 120 °C.



(a)

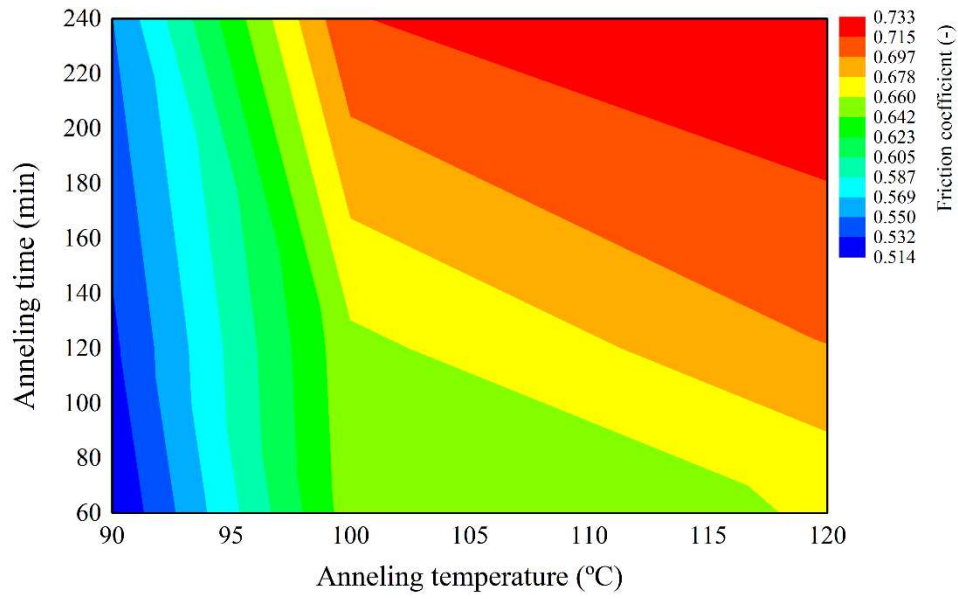


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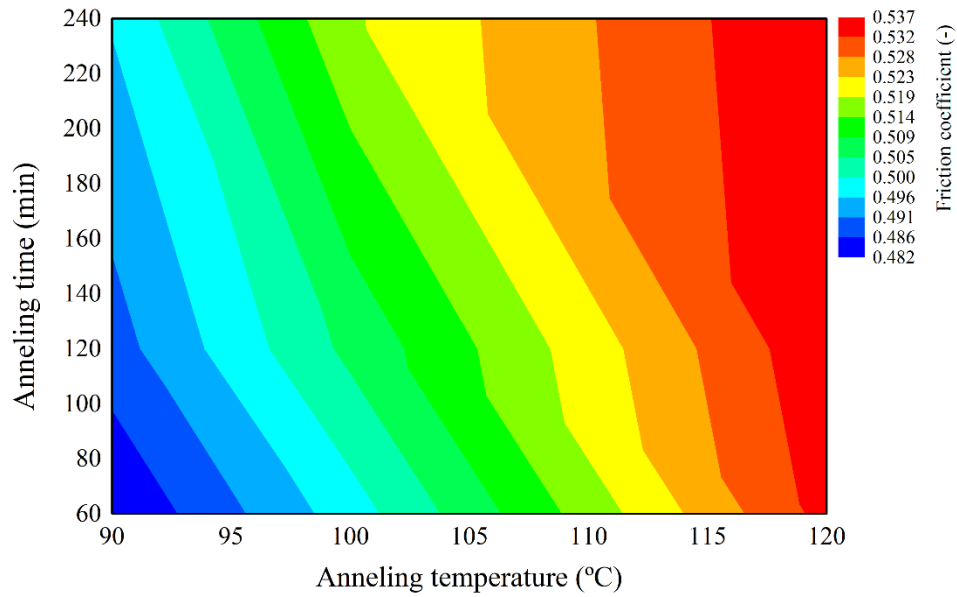


(c)

Figure 3. Evolution of the friction coefficient as a function of sliding distance for graphene-reinforced PLA specimens annealed for 60, 120, and 240 min at: (a) 90 °C, (b) 100 °C, and (c) 120 °C.



(a)



(b)

Figure 4. Effect of annealing temperature and time on the friction coefficient of FFF-printed (a) neat PLA and (b) graphene-reinforced PLA

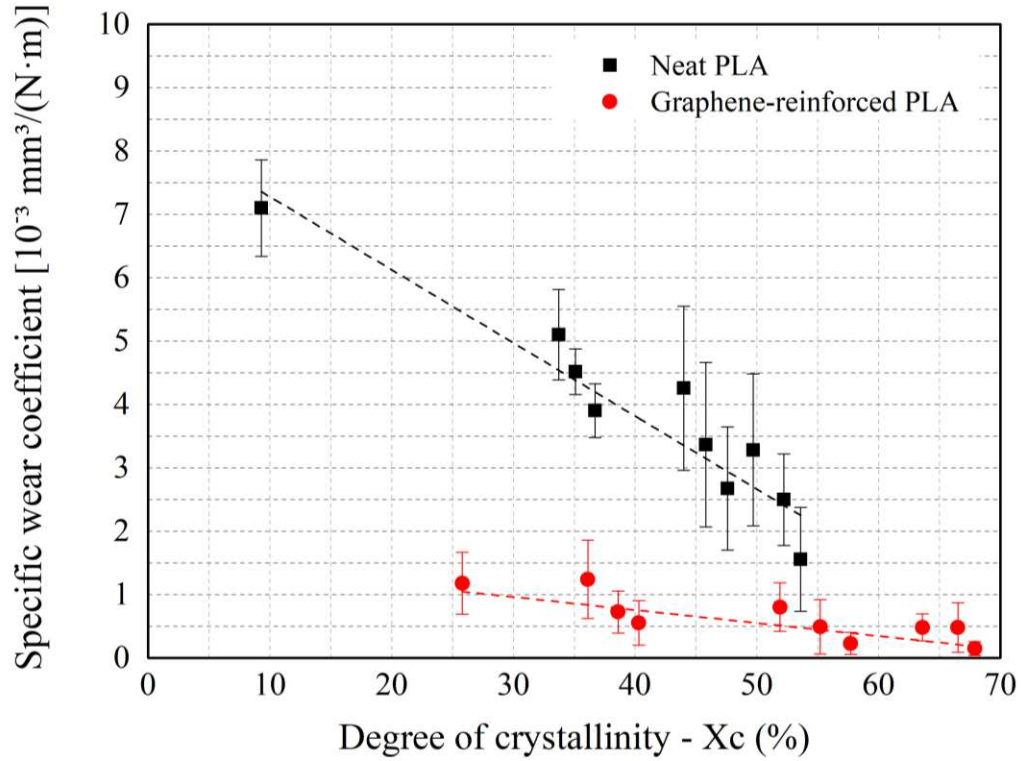
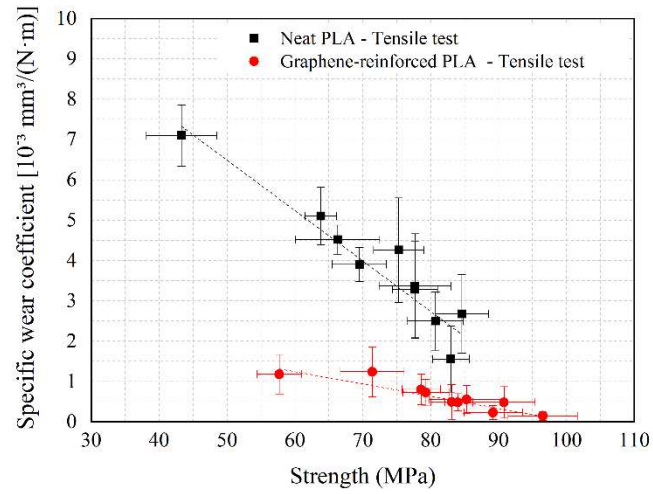
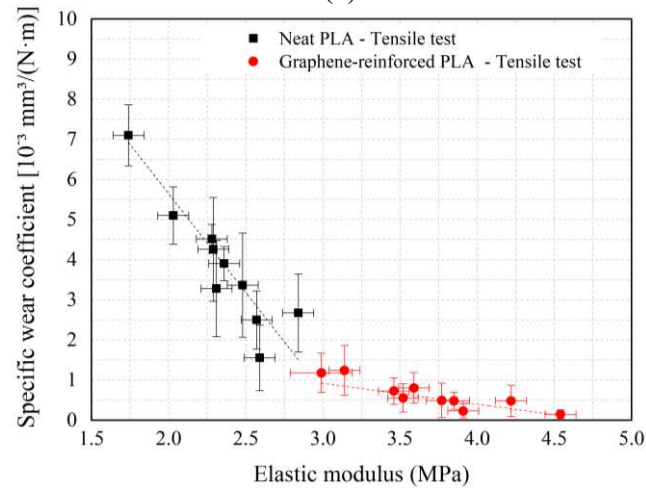


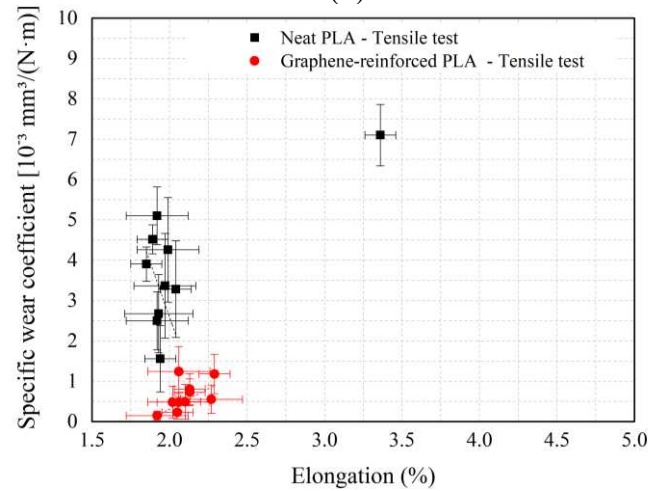
Figure 5. Variation of the specific wear coefficient as a function of the degree of crystallinity for FFF-printed neat PLA and graphene-reinforced PLA. Dashed lines represent the linear regression fits, and error bars indicate the standard deviations.



(a)



(b)



(c)

Figure 6. Variation of the specific wear coefficient as a function of the tensile properties of FFF-printed neat PLA and graphene-reinforced PLA: (a) tensile strength, (b) tensile elastic modulus, and (c) strain at break.

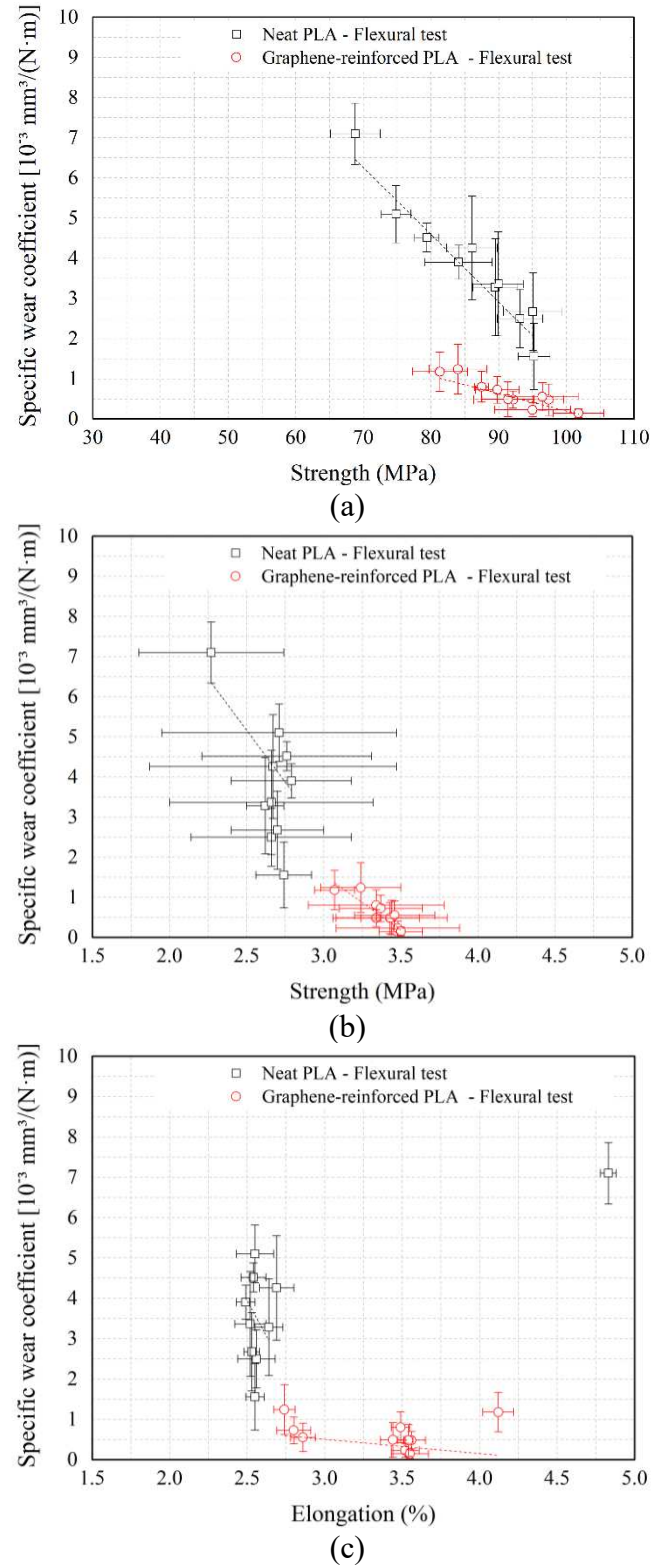


Figure 7. Variation of the specific wear coefficient as a function of the flexural properties of FFF-printed neat PLA and graphene-reinforced PLA: (a) flexural strength, (b) flexural modulus, and (c) flexural strain.

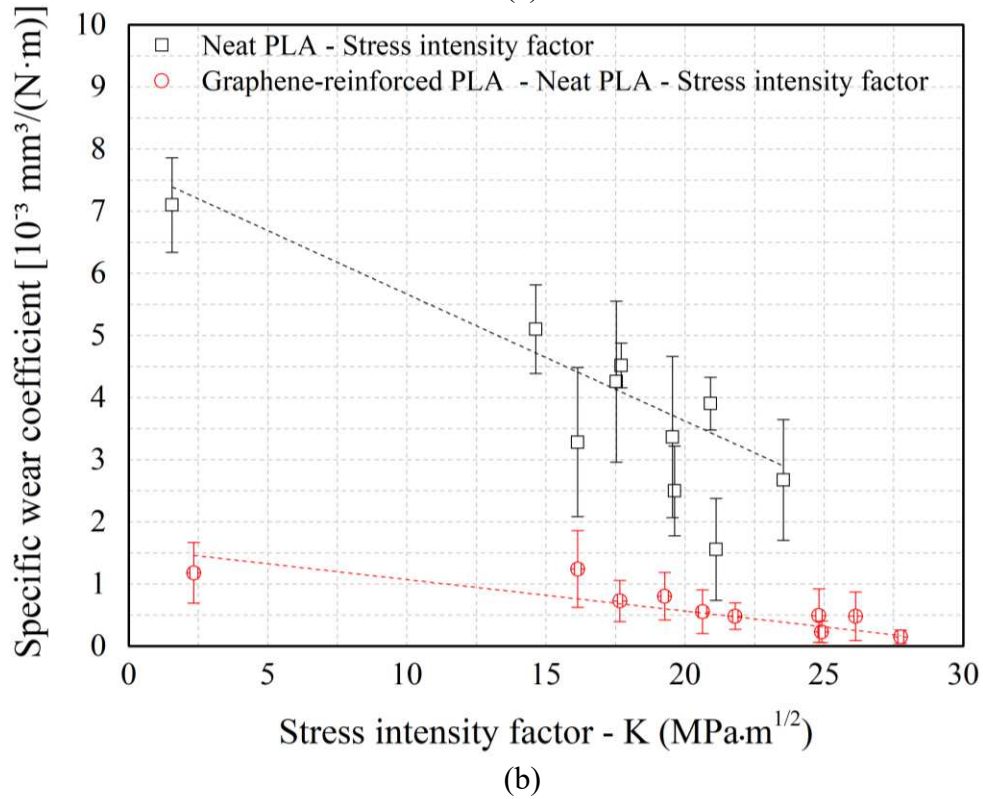
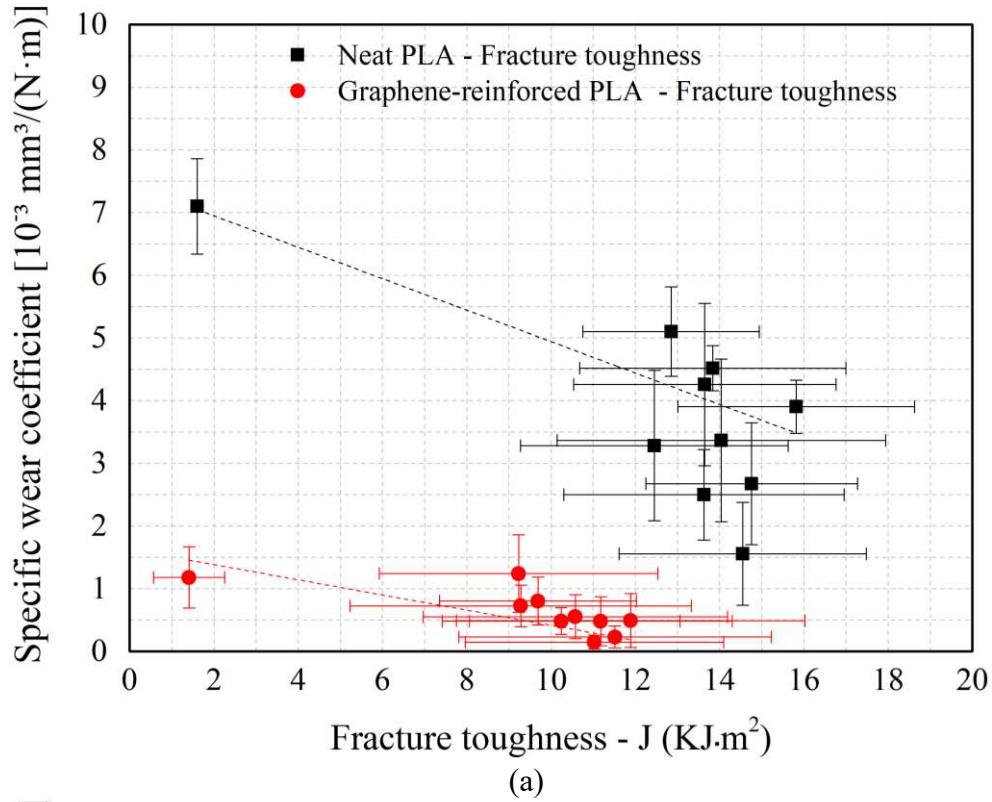


Figure 8. Variation of the specific wear coefficient as a function of the fracture-mechanics parameters of FFF-printed neat PLA and graphene-reinforced PLA: (a) J-integral and (b) stress intensity factor.

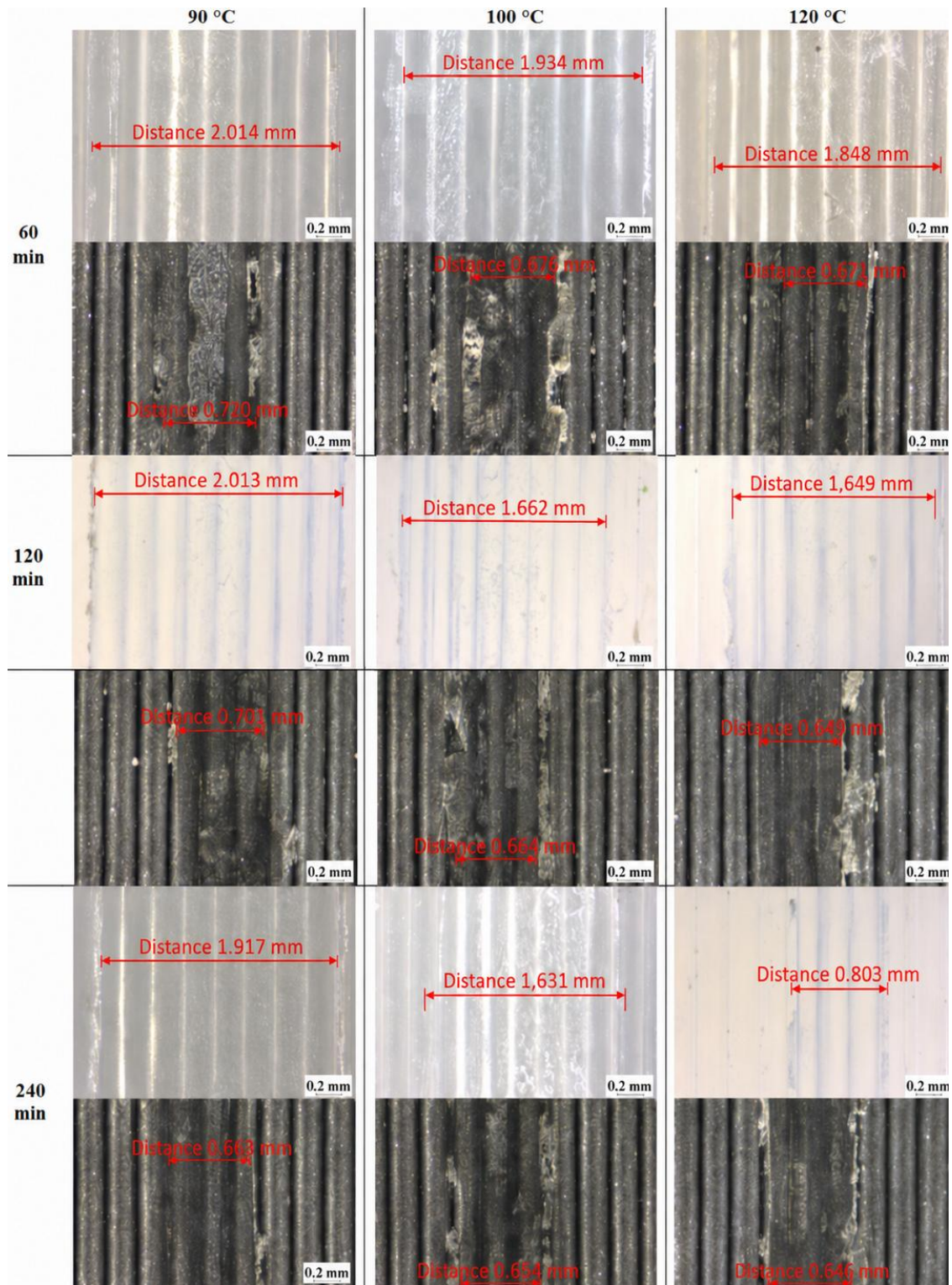
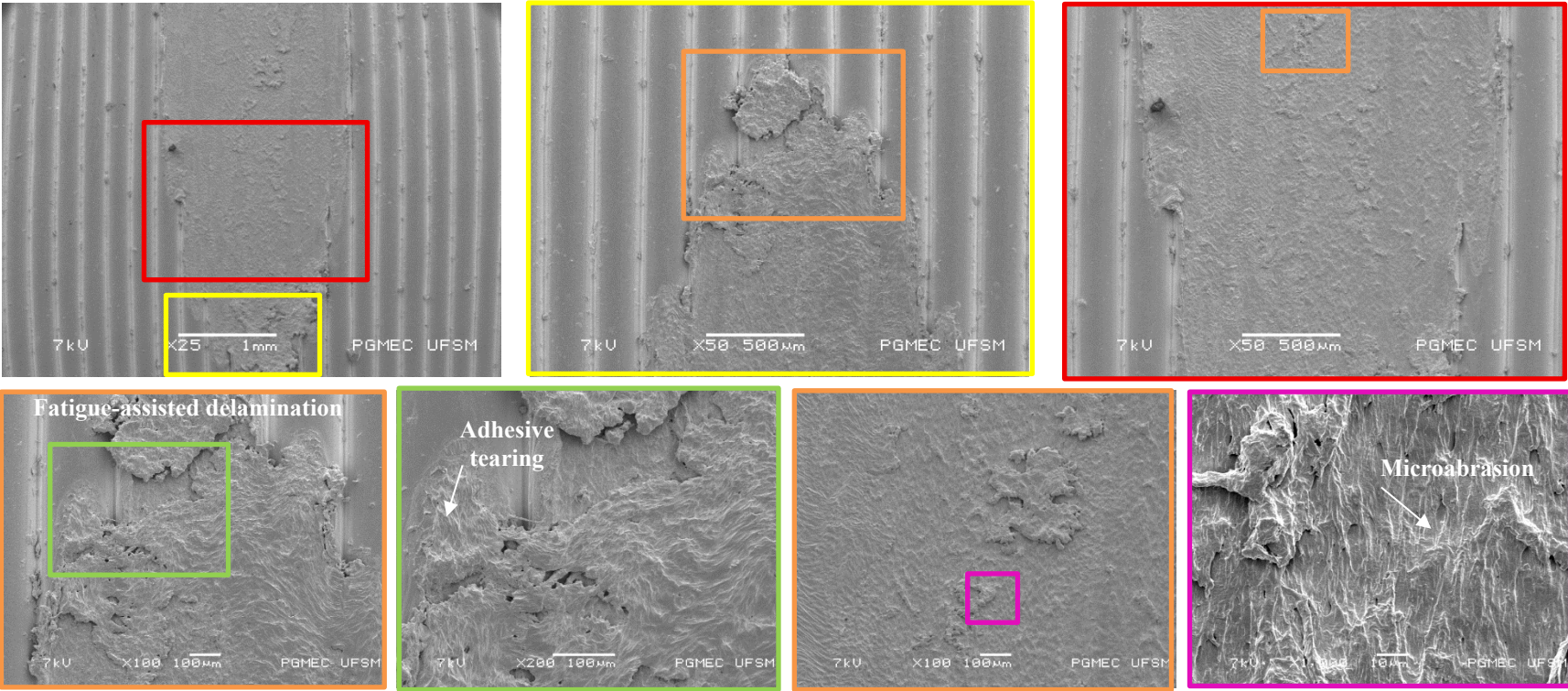
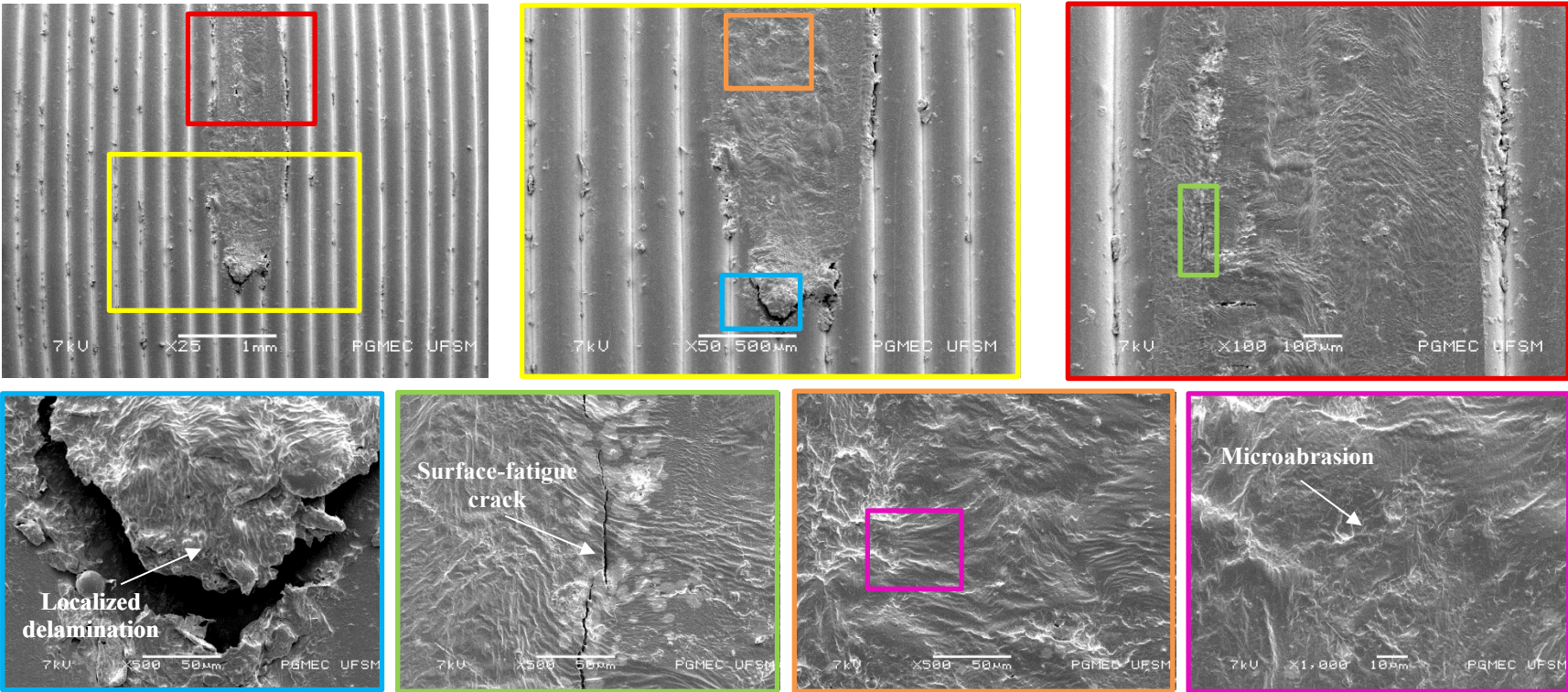


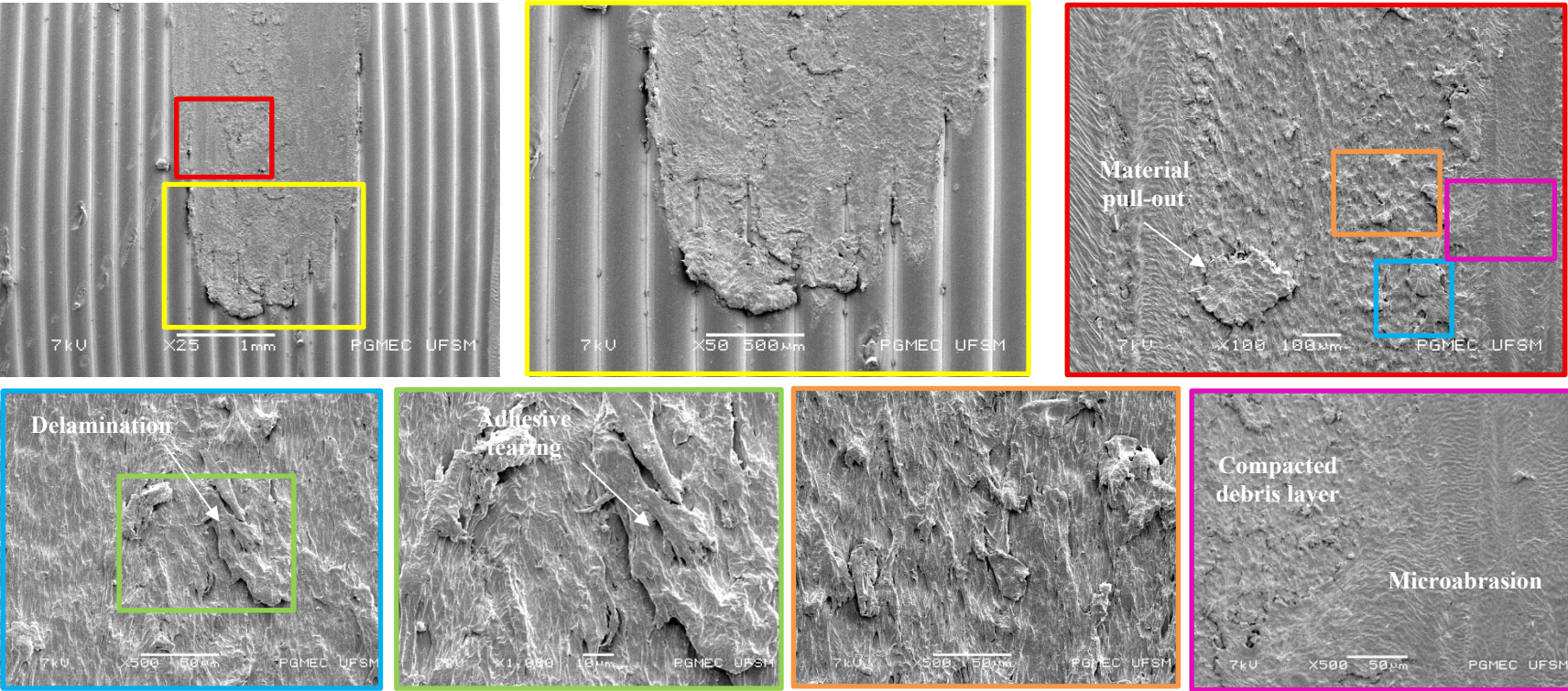
Figure 9. Stereomicrographs of the wear tracks formed on FFF-printed neat PLA and graphene-reinforced PLA specimens annealed for 60, 120, and 240 min at 90, 100, and 120 °C. Light-colored specimens correspond to neat PLA, whereas dark-colored specimens correspond to graphene-reinforced PLA. Red arrows indicate the wear-track width.



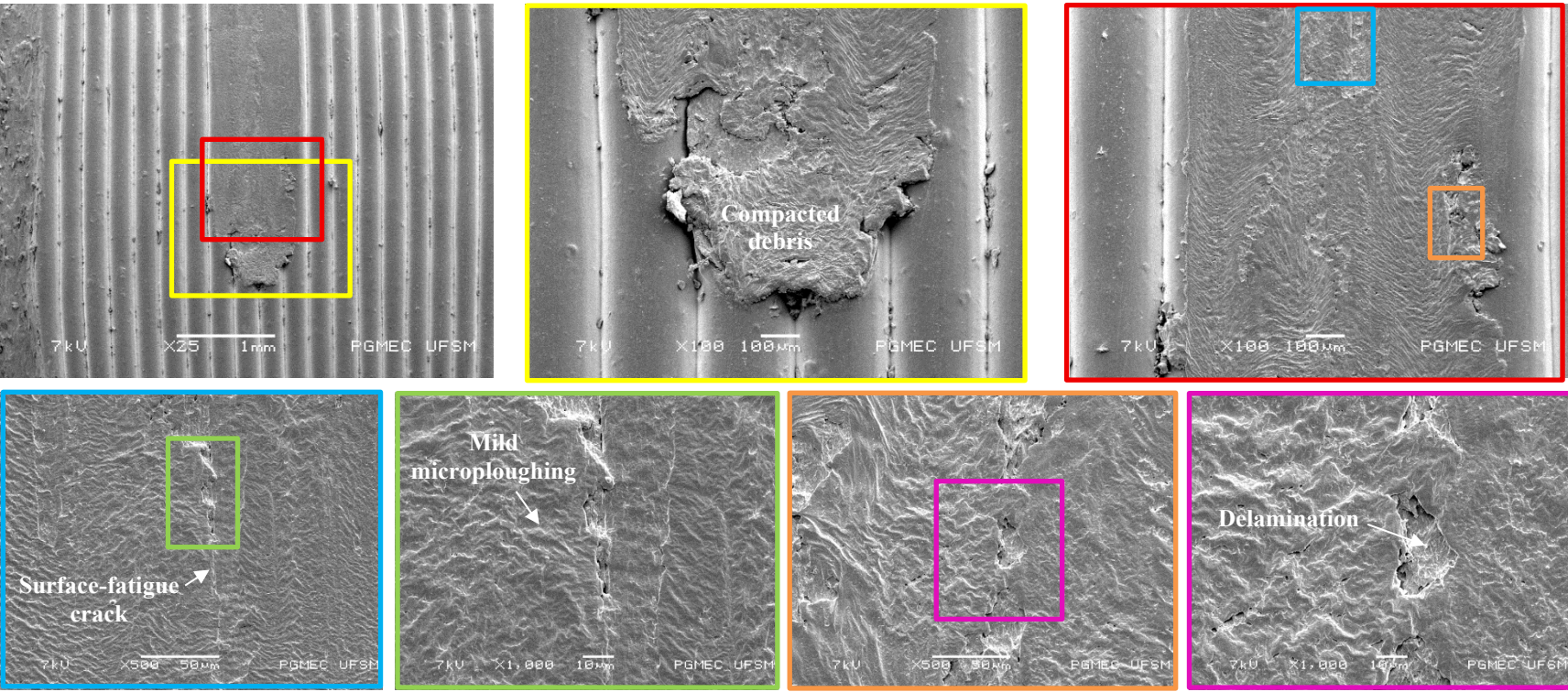
(a1)



(a2)



(b1)



(b2)

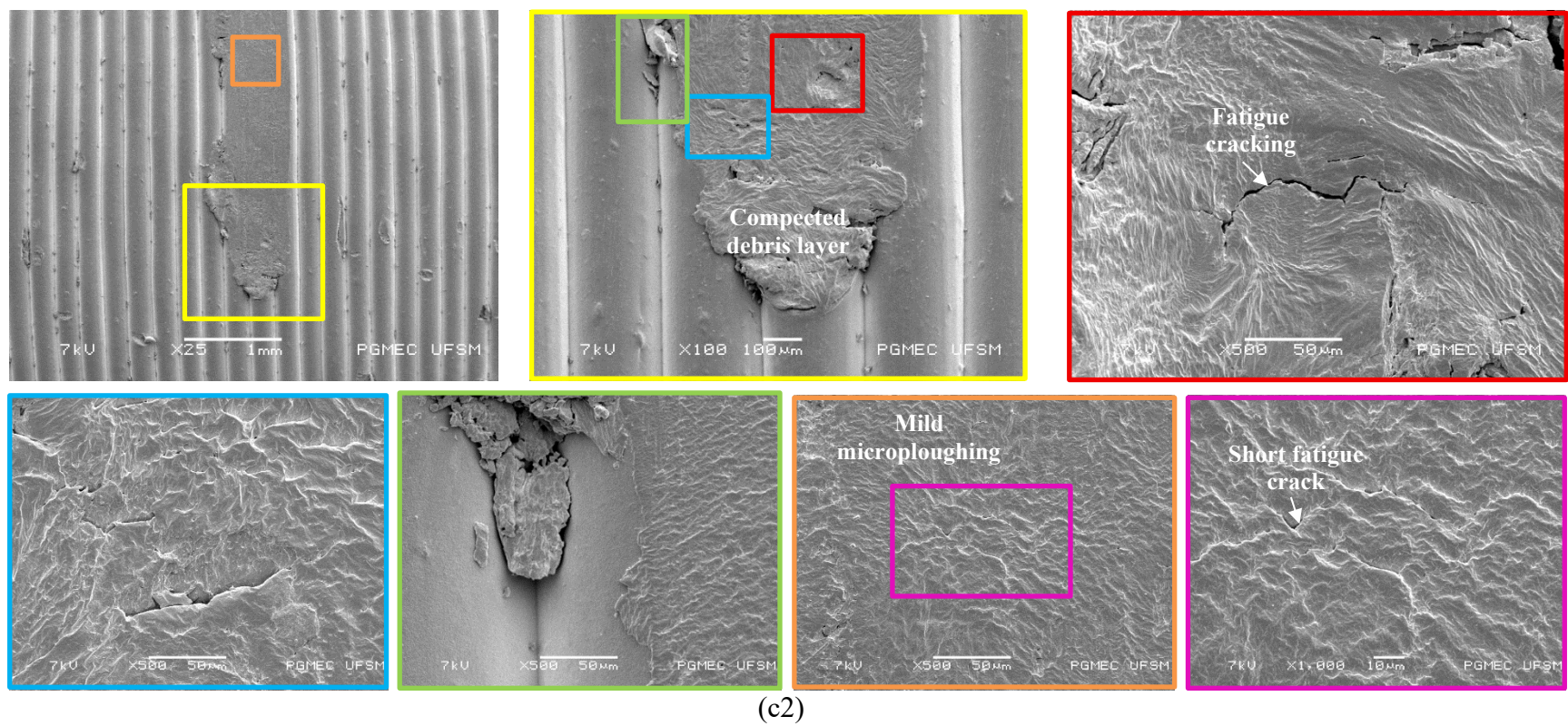
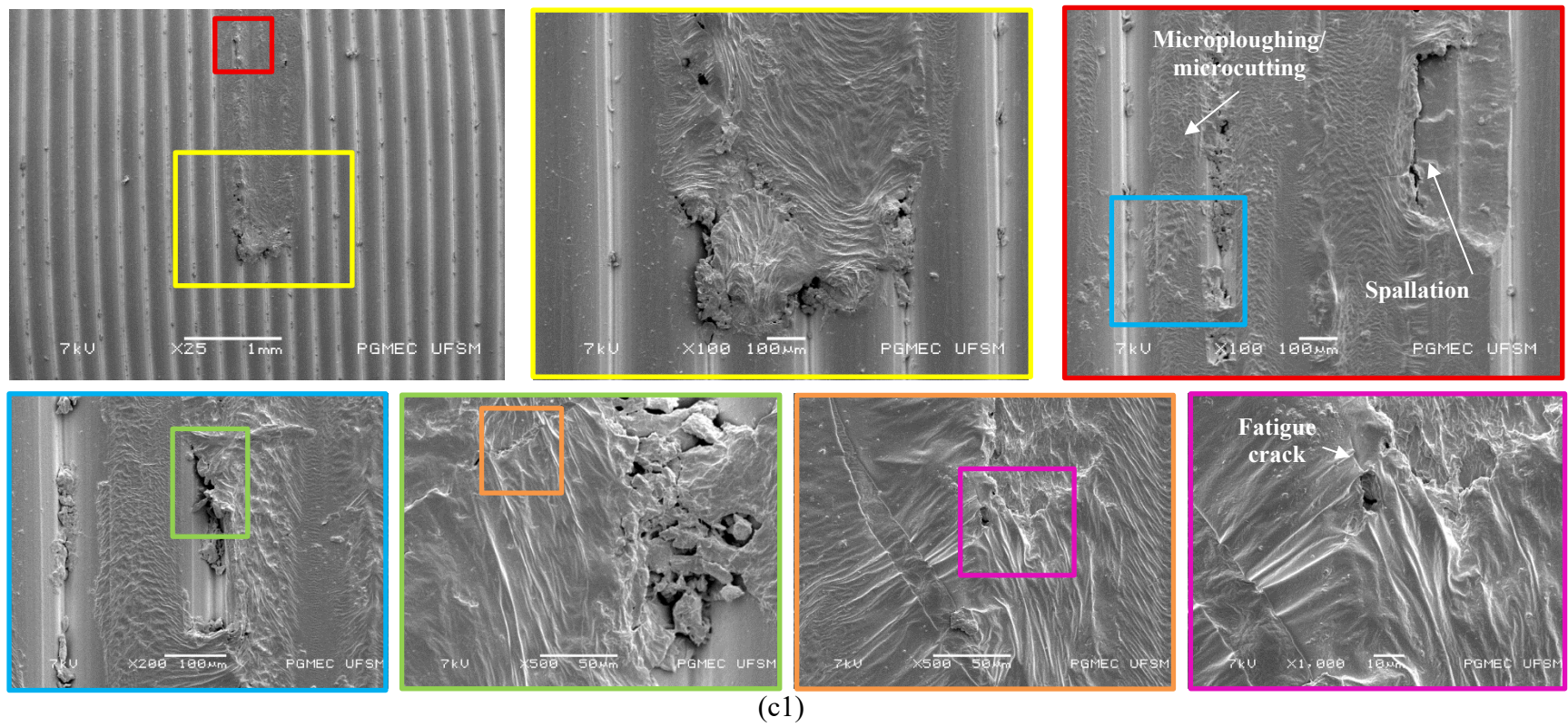


Figure 10. SEM montages of the wear-track regions of FFF-printed neat PLA and graphene-reinforced PLA after annealing under selected conditions: (a1) neat PLA and (a2) graphene-reinforced PLA annealed at 90 °C for 60 min; (b1) neat PLA and (b2) graphene-reinforced PLA annealed at 100 °C for 120 min; and (c1) neat PLA and (c2) graphene-reinforced PLA annealed at 120 °C for 240 min.