Investigation of The Use of PEG as a Compatibilizer in Nanocellulose Reinforced PLA Composites

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Poly(lactic acid) (PLA) is a widely used biodegradable polymer due to its renewable origin and favorable environmental properties. However, its low ductility and impact resistance limit its use in more demanding applications. In this context, this study investigated the formulation of PLA composites with a masterbatch containing natural trichomes, cellulose nanofibrils (NFC), and polyethylene glycol (PEG), with the aim of improving the interfacial compatibility and mechanical properties of the material. The literature shows that NFC can reinforce the polymer matrix, but its dispersion is hampered by the difference in polarity with PLA, making it necessary to use compatibilizers such as PEG. The experimental results indicated that formulations with balanced NFC and PEG contents showed significant improvements in mechanical strength and deformation at break, especially those that employed the PEG masterbatch (MB) strategy. The proposed approach proved to be effective and sustainable, expanding the potential application of PLA in sectors that demand higher performance, such as technical packaging, biodegradable films, and structural components from renewable sources.
Keywords: PLA. Cellulose Nanofibrils. PEG. Compatibilization. Biocomposites.

Growing environmental concern over the accumulation of non-biodegradable plastic waste has led to the development of sustainable alternatives, such as polymers of renewable origin. Among these, poly(lactic acid) (PLA) stands out because it is biodegradable, biocompatible and comes from natural sources such as corn starch or sugar cane. However, PLA has limited mechanical properties, such as low impact resistance and reduced ductility, which restricts its application in more demanding sectors [1].

In order to overcome these limitations, the incorporation of natural trichome reinforcements, such as cellulose nanofibrils (NFC), has proved to be a promising alternative. Due to its high stiffness, high aspect ratio and abundance of hydroxyl groups, NFC acts as a structural reinforcement and can significantly improve the modulus of elasticity and tensile strength of composites [2,3]. However, the presence of these hydroxyl groups also gives NFC its hydrophilic character, which makes it difficult to disperse it homogeneously in hydrophobic matrices such as PLA [4].

In this scenario, the use of compatibilizing additives such as poly(ethylene glycol) (PEG) has become an effective strategy. PEG can interact with the surface of the NFC through hydrogen bonds, promoting surface modification of the nanofiber and facilitating its incorporation into the polymer matrix. In addition, PEG acts as a plasticizer, reducing the crystallinity of PLA and increasing its ductility [5].

The study by Cailloux and colleagues (2019) [6] demonstrated that levels of up to 20% PEG in wet suspension can be used in NFC pretreatment, favoring its individualization. This procedure improves nanofibril dispersion and contributes to greater compatibility with the PLA matrix, optimizing the composite's performance. The study carried out by, Clarkson and colleagues (2020) [7] used lower concentrations, such as 5% by weight, also with satisfactory results. This data indicates a wide range of possible PEG contents to be investigated, with direct impact on the structural and functional properties of the composite.

Therefore, this work proposes a PLA composite formulation containing a masterbatch with

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natural trichomes, NFC, and PEG, with the aim of evaluating the efficiency of PEG as a compatibilizing agent and plasticizer, promoting better nanofiller dispersion and improving mechanical performance. The choice of this composition is based on several pieces of evidence in the literature [2,8,9], which point to the synergistic potential between natural reinforcements and plasticizers in biodegradable matrices.

Materials and Methods

The composites were prepared by melt mixing PLA (Natureworks 3D850), masterbatch (MB: PLA/5wt% trichome), nanofibrils (manufactured by TRL9) and PEG (PEG-600) (Table 1 and 2), using co-rotating twin-screw extruder (DR.16.40.AX from AX Plásticos), speed 135 rpm, temperature range 150 to 190. Followed by filament production using Filmaq 3D single-screw extruder, speed 32 rpm, temperature 190ºC. And 3D printing (Prusa Research MK3S+ 3D) to produce tensile test specimens.

Table 1. Formulations without added PEG.

Formulation	PLA	Masterbatch	NFC
F1	100.0%	–	–
F2	99.5%	0.5%	–
F3	99.5%	–	0.5%
F4	99.0%	0.5%	0.5%
F5	98.5%	0.5%	1.0%
F6	97.5%	0.5%	2.0%

Table 2. Formulations with added PEG.

Formulation	PLA	Masterbatch	NFC	PEG
F1	97.0%	–	–	3.0%
F2	96.5%	0.5%	–	3.0%
F3	96.5%	–	0.5%	3.0%
F4	96.0%	0.5%	0.5%	3.0%
F5	95.5%	0.5%	1.0%	3.0%
F6	94.5%	0.5%	2.0%	3.0%

The formulation specimens were prepared in accordance with ASTM D638, type IV and were tested on an EMIC model DL200MF universal testing machine with a 2 kN load cell, speed test 5 mm/min, 5 replicates for each formulation. The properties analyzed were tension at maximum force (MPa), specific deformation at break (%) and modulus of elasticity (MPa). Morphological analyses were performed using digital optical microscopy (Nova Digital) to evaluate the fracture surface of the fractured tensile specimens.

Data from the tensile tests were analyzed by ANOVA, followed by the Tukey test ($p<0.05$) to compare means, using Statistica 7.0 software.

Results and Discussion

A Prusa Research MK3S+ 3D printer was used to fabricate the specimens (Figure 2). This method ensured precise control of the sample geometry, enabling consistent and reproducible preparation for mechanical characterization. The printing process also demonstrated good processability of the formulations, especially those containing PEG, which contributed to smoother extrusion and an improved surface finish.

Figures 3, 4, and 5 display the results of the tensile tests conducted on the different formulations, representing, respectively, the stress at maximum force, the modulus of elasticity, and the deformation at rupture.

Analysis of the ultimate tensile strength results presented in Figure 3 allows for a comprehensive assessment of the influence of NFC, masterbatch (MB), and PEG on the mechanical performance of PLA-based composites. The formulations with and without PEG show that the isolated incorporation of masterbatch or cellulose nanofibrils does not significantly increase tensile strength. This result highlights the limitations of reinforcement efficiency when additives are used individually, likely due to poor interfacial adhesion and inadequate dispersion within the hydrophobic PLA matrix, which are well-documented challenges in

Figure 1. Description of steps.

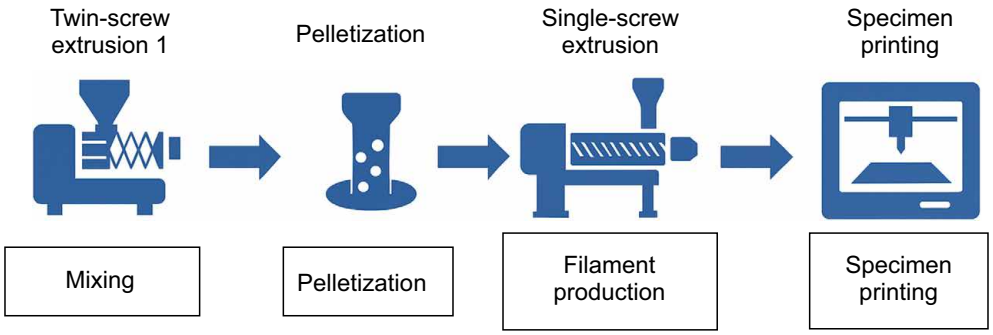


Figure 2. Tensile test specimen of composites obtained by 3D printing.

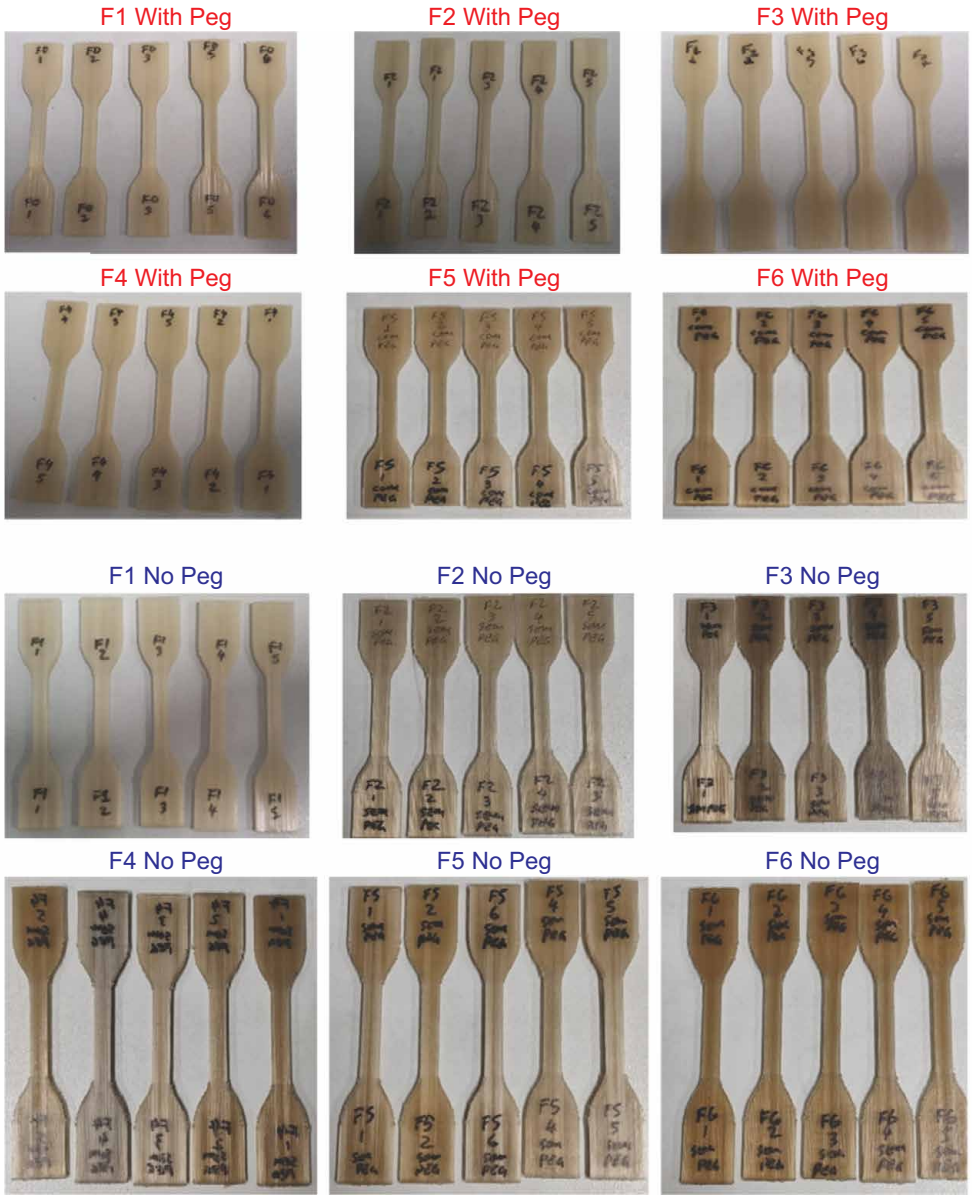


Figure 3. Stress at maximum strength of PLA formulations with and without PEG.

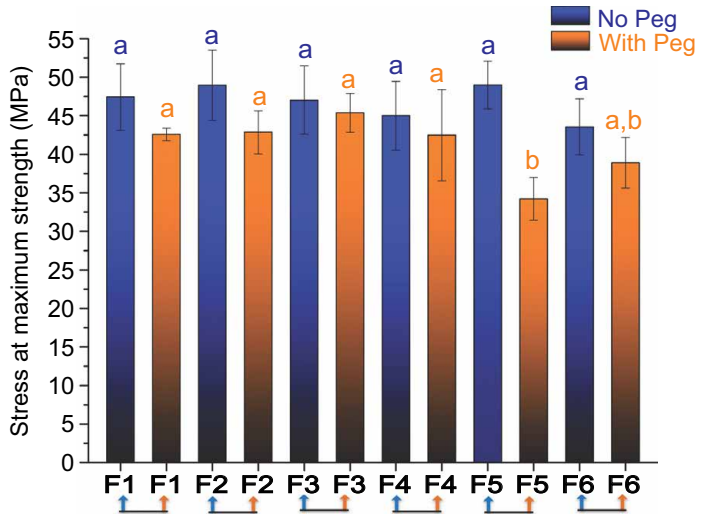


Figure 4. Modulus of elasticity of PLA formulations with and without PEG.

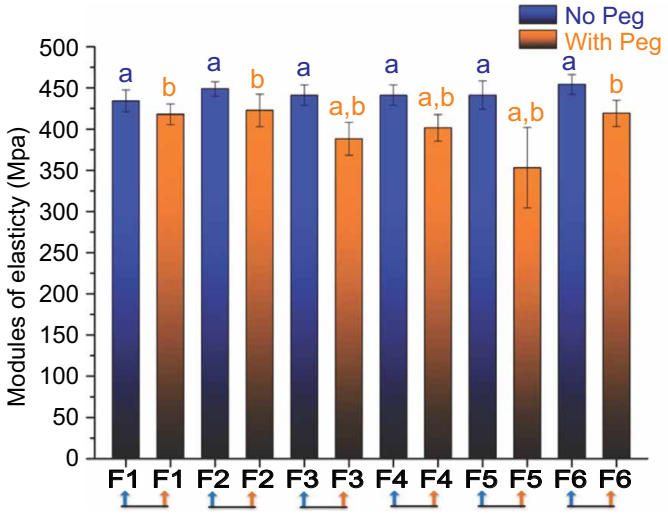
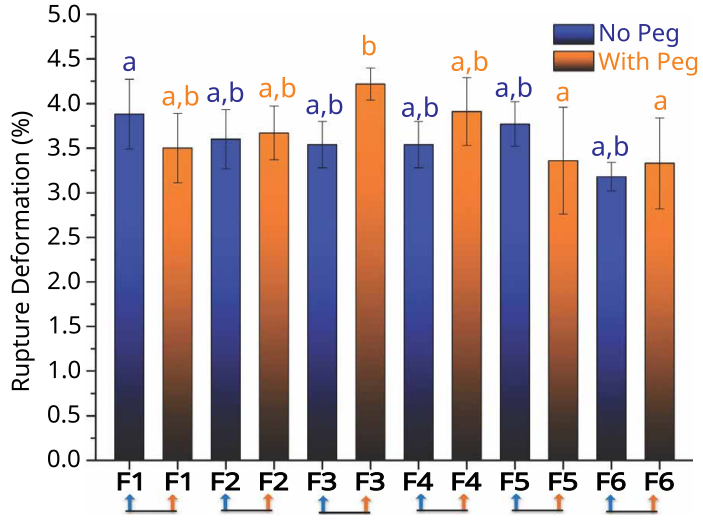


Figure 5. Deformation at break of PLA formulations with and without PEG.



the processing of PLA/nanocellulose composites [10,11].

Formulations F5 and F6, which contain higher NFC contents (1 and 2%, respectively), exhibit a marked reduction in tensile strength, particularly in the presence of PEG. This may be attributed to the difficulty in maintaining dispersion at high NFC loadings, leading to agglomeration and localized strength concentrations that compromise mechanical integrity [7-12].

Interestingly, the PEG-free F6 formulation partially reverses this trend, exhibiting higher strength than its PEG-containing counterpart, as evidenced by the Tukey test, specifically, F5 and F6 with PEG share the Tukey subindex "b," indicating statistically distinct and lower mean values compared to the other samples. This suggests that eliminating PEG in high-NFC systems helps preserve matrix strength and allows the reinforcing effects of nanocellulose to manifest more effectively, provided dispersion is sufficiently controlled [13].

The tensile strength results highlight the critical role of the balance between additive content, dispersion quality, and interfacial compatibility in PLA-based composites. These findings reaffirm the importance of fine-tuning composite formulations to maximize mechanical performance, as corroborated by previous studies on biobased nanocomposites [6-14].

The modulus of elasticity, or Young's modulus, is a key parameter that reflects a material's capacity to withstand deformation when subjected to applied stress. As shown in Figure 4, the results clearly demonstrate the effects of NFC content, the presence of PEG, and the synergy between additives on the stiffness of PLA composites.

Formulation F6 (PLA + 2% NFC, no PEG) exhibited the highest Young modulus (454.42 MPa), highlighting the reinforcing effect of nanocellulose at higher concentrations in the absence of plasticizer. This finding is in line with previous studies indicating that well-dispersed NFCs increase the brittleness of PLA due to their high aspect ratio and strong hydrogen bonding network [6,7]. The absence of PEG, in this case, preserves the interfacial tension between the matrix and the nanofibrils, maximizing the reinforcement potential. Similarly, formulation F2 (PLA + MB, no NFC) achieved a high modulus (449.02 MPa), indicating that the functionalized additives on the masterbatch surface can promote polymer chain alignment or reduce segmental mobility, thus increasing the difficulty. However, the addition of PEG to this formulation (F2 with PEG) reduces the modulus to 422.76 MPa, which is consistent with the known plasticizing behavior of PEG, which increases chain mobility and reduces intermolecular interactions [6-16].

The effect of PEG becomes even more pronounced in formulation F5 (PLA + MB + 1% NFC), where the modulus dropped from 441.42 MPa to 353.28 MPa after PEG incorporation. This sharp drop reveals the challenge of achieving a favorable balance between reinforcement and plasticization. It is likely that, at this NFC concentration, PEG not only interferes with NFC dispersion but also increases chain flexibility to the point of weakening the composite structure, a characteristic also observed in other NFC-PLA systems with poor interfacial control [13,14].

On the other hand, formulation F3 (PLA + NFC, no masterbatch) in the presence of PEG resulted in one of the lowest impairment values (388.37 MPa), confirming that, in the absence of compatibilizers, PEG can impair reinforcement efficiency. The lack of interfacial adhesion PLA/NFC, combined with the mobility-enhancing effect of PEG, results in deformable and less rigid composite behavior, also reported by Cheng and colleagues (2015) [15] in NFC-reinforced systems without surface treatment.

In summary, the analysis of the elastic modulus between the formulations highlights the critical importance of the balance between reinforcement content, compatibilization, and plasticization. PEG reduces rigidity, especially when interfacial interactions are weak.

Strain at break is a key indicator of ductility, reflecting a material's ability to undergo plastic

deformation before fracture. As illustrated in Figure 5, significant differences in deformation behavior were observed between PLA-based formulations, influenced by the presence of reinforcing agents and plasticizers.

With the isolated addition of masterbatch (F2) or NFC (F3), a reduction in deformation is observed, especially in the absence of PEG. This can be attributed to the increased rigidity and brittleness of the matrix, since the additives, when incorporated, hinder the transfer of polymer chains. When there is no compatibility or specific dispersion, there is a greater formation of stress concentration points that anticipate material rupture. The presence of PEG promotes significant changes. In formulation F3 with PEG, for example, there is a significant increase in strain at break, decreasing the likelihood that PEG acted effectively as a plasticizer, increasing chain mobility, and conferring greater flexibility to the compound. This behavior is consistent with previous studies showing that PEG reduces intermolecular interactions in PLA, increasing ductility [17].

In formulation F3 with PEG, the strain at break increased markedly, confirming the effective plasticizing role of PEG. This result is in line with literature reports that PEG can significantly increase ductility by disrupting PLA's intermolecular interactions and increasing polymer chain flexibility [6,7]. This formulation achieved the highest ductility among all samples, demonstrating the synergistic effect of plasticization in an unreinforced matrix.

Formulations F5 and F6, which incorporated 1 and 2% NFC, respectively, revealed interesting contrasts. F5 with PEG showed increased strain compared to its PEG-free counterpart, although to a lesser extent than F3. This suggests that the plasticizing effect of PEG is partially impaired by the increased NFC content. For F6 with PEG, the strain dropped to the lowest value among all samples. This counterproductive result is consistent with the appearance of reinforcement supersaturation, where high NFC loadings, especially when poorly dispersed, create microstructural defects that act as failure initiation sites, impairing ductility [7-17].

Interestingly, F6 without PEG exhibited greater strain than F6 with PEG. Although counterintuitive, this may reflect the formation of a more consistent internal NFC network in the absence of PEG, providing a better monetary response before rupture. PEG, in this case, may have disrupted this network, leading to premature failure due to microphase separation or void formation during deformation, a phenomenon observed in polymer nanocomposites with low matrix-filler compatibility [13-17].

Overall, these findings demonstrate that ductility in PLA composites is governed by a delicate balance between plasticization and reinforcement. While PEG enhances flexibility, its effect is significantly influenced by the type, concentration, and dispersion of reinforcing agents. The most ductile behavior was observed in F3 with PEG, while the most brittle occurred in F6 with PEG, highlighting the critical role of formulation design in optimizing mechanical performance.

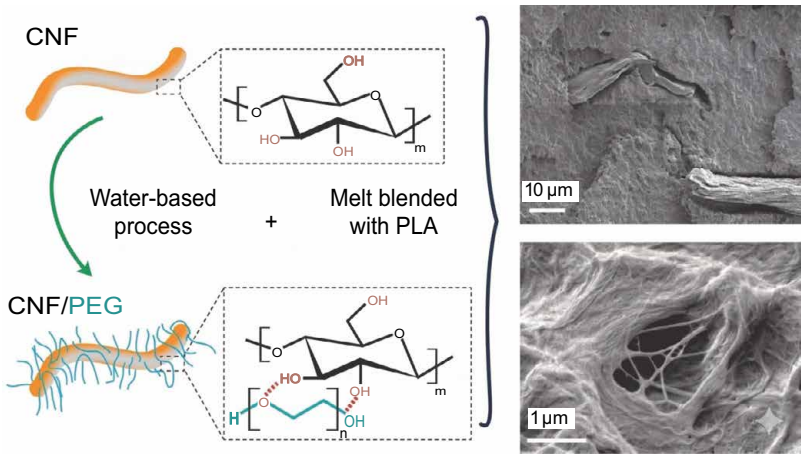
Figure 6 shows a schematic representation of hydrogen bonding interactions PEG-NFC, promoting a well-entangled reinforcement network.

Optical microscopy examination of the fractured surfaces of tensile test specimens (Figure 7) reveals that the samples containing PEG exhibit a more ductile fracture morphology, characterized by stretched and deformed regions, whereas the samples without PEG display a comparatively brittle fracture appearance.

The melt-processing of cellulose nanofibril/ polylactide bionanocomposites via a sustainable polyethylene glycol-based carrier system [6] (Figure 6), it can be concluded that the effective dispersion of NFC in PLA through the incorporation of PEG as a carrier was critical to the mechanical and structural performance of the developed composites.

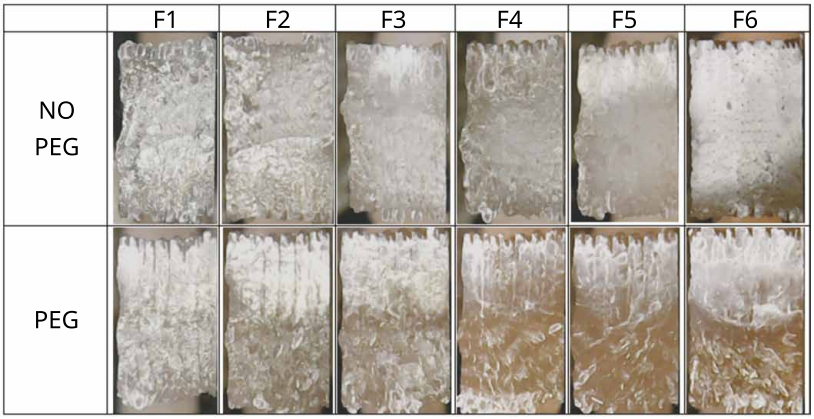
The analyses showed that moderate levels of PEG enable the formation of oxygen bonds

Figure 6. Hydrogen bonding interactions PEG-NFC.



Adapted from Cailloux and colleagues (2019) [6].

Figure 7. Optical microscopy of fractured surfaces of tensile test specimens.



between the NFC and the polymer matrix, which facilitates the dispersion of the nanofibrils and prevents their agglomeration, one of the greatest challenges in the formulation of melt-formed nanocomposites. Formulations F3 and F4, with balanced NFC and PEG contents, stood out for presenting good strength and deformation, demonstrating a well-structured network. On the other hand, formulations with higher NFC concentrations (such as F5 and F6 with PEG) showed poor performance, likely due to network saturation and excessive plasticization.

Thus, the study confirms that the use of masterbatch and NFC with PEG is an effective strategy for improving compatibility and dispersion in PLA, optimizing the material's mechanical properties. This system represents a viable advancement for the production of biocomposites with balanced performance, aligning sustainability, processability, and structural efficiency.

Conclusion

This study demonstrated the effectiveness of using polyethylene glycol (PEG) as a carrier for the dispersion of cellulose nanofibrils (NFC) in a polylactide (PLA) matrix, enabling the production of bionanocomposites through melt processing. The masterbatch formulation with PEG was designed to be strategic in promoting intermolecular interactions, such as protective bonds, which favored compatibility between components and minimized the typical challenges of NFC agglomeration.

The mechanical results indicated that formulations with moderate NFC and PEG contents, especially F3 and F4, demonstrated the best balance between strength, damage, and deformation, revealing optimized performance for applications that unlock flexibility and structural integrity. Conversely, excess NFC and plasticizer compromised performance due to agglomeration and excessive plasticization, highlighting the need of composition control.

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