High-Performance PET Composites Reinforced with Hybrid Nanocellulose–Silica Nanorods

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1. Abstract

Current reinforcing materials used in high-performance composites are typically carbon- or glass fibre-based. These materials are not biodegradable and end up in landfills or require extensive energy to recycle, slowing the transition to a sustainable circular economy. Bio-based and biodegradable composite fillers are of growing interest but often suffer from issues surrounding matrix incompatibility and thermal instability. In this work, nanocellulose-templated silica nanorods (SiNRs) are introduced as a hybrid bio-based filler for composites, featuring tunable matrix compatibility while maintaining high mechanical performance and improved sustainability and biocompatibility. Material loadings up to 5 wt% of SiNRs were dispersed in solutions with polyethylene terephthalate (PET), spin-coated onto Teflon slides to create thin films and cut into strips for tensile testing. An increase in elastic modulus of up to 73% and a yield strength (YS) improvement exceeding 25% was observed. In addition, the thermal degradation behaviour of the polymer was not greatly affected or diminished by the filler. These results demonstrate that high mechanical performance can be achieved using SiNRs at very low loadings, highlighting their potential as a more sustainable reinforcing filler for polymer composites.

2. Introduction

Polyethylene terephthalate (PET) is one of the most commonly used plastics today, seen in numerous industries such as packaging, textiles, sporting goods, automotive, and construction [1]. One benefit of using PET is that it is easily recyclable once separated, however recycled PET typically lacks the mechanical performance of its raw form [2]. Thus, in applications where enhanced mechanical properties are required, PET is often reinforced with filler materials to meet those demands. Current reinforcing additives for polymers, such as carbon and glass fibres, are derived from non-renewable sources, face issues with recyclability, and are commonly directed to landfill at end-of-life [3]. The production of these materials requires energy-intensive processes and results in significant greenhouse gas (GHG) emissions [4, 5]. Glass fibre production creates approximately 2,000 CO_2eq/tonne of fibre, while carbon fibre produces between 8,000 – 29,500

CO_2eq/tonne of fibre [6, 7]. Comparatively, natural fibres such as flax, hemp, jute or kenaf produce less than 1,000 CO_2eq/tonne of fibre and boast the benefit of being biodegradable due to their organic nature [6].

Bio-based alternatives are of growing interest as composite fillers but often lack the durability of conventional filler materials and suffer from other issues such as incompatibility with polymer matrices, poor moisture resistance, and variability due to their natural sourcing [8, 9]. In this work, bio-based nanocellulose-templated silica nanorods (SiNRs) are presented as a promising alternative filler for PET composites that may help address some of these challenges. SiNRs are hybrid rod-shaped nanoparticles with an organic cellulose nanocrystal (CNC) core and an inorganic silica shell. The particles are partially renewable in their nature due to their CNC core, with a chemically benign and biocompatible silica shell. On their own, CNCs possess excellent mechanical properties but lack thermal stability and show incompatibility when mixing with hydrophobic solvents and polymer matrices due to their hydrophilic nature [10, 11]. By protecting the CNC core with a coating of silica, the thermal stability of the CNC template is vastly improved, as was demonstrated in the work by Jankovic et al. [12].

To address the issue of matrix incompatibility, SiNRs can be easily modified due to their tunable surface chemistry to possess hydrophobic, oleophobic, or even anti-bacterial properties if desired [13, 14]. The rod-like geometry of SiNRs provides a stronger reinforcing effect than typical spherical silica nano particles – acting like glass fibres, but at the nanoscale. Additionally, due to their high aspect ratio, lower loadings of SiNRs are required to achieve the same mechanical performance as typical fillers. Whereas a traditional composite might need 20 wt% glass fibre to reach a certain elastic modulus, a nanocomposite might only need 1.5 wt% filler to achieve the same performance [15, 16]. Increased thermal stability will aid the nanomaterials during thermal extrusion processing, which is one of the more common industrial production practices. However, solvent casting is still relevant to the scientific community and will be explored in this study [17].

SiNRs are produced by growing silica on the surface of cellulose nanocrystals (CNCs) via the Stöber process in solution with tetraethyl orthosilicate (TEOS) and ammonium hydroxide. CNCs are derived from cellulose found in plant cell walls, which is typically processed from wood pulp or other agricultural by-products through acid hydrolysis. The production of TEOS depends on silicon derived from quartz, which is typically produced through a high-temperature carbothermic reduction around 2000°C [10]. This process is highly energy-intensive and relies on non-renewable sources. However, Nguyen et al. have devised a method to produce TEOS from rice husks. While their method is still new and facing limitations, such as the excess usage of ethanol, initial life cycle assessment (LCA) calculations are promising [11]. Additionally, instead of using petroleum-based polymers like PET, other bio-based and/or biodegradable polymers derived from polysaccharides (cellulose, starch, chitin, etc.) or polylactic acid (PLA) could be explored to make the entire composite from renewable or more sustainable materials.

End-of-life (EoL) management remains a significant challenge for nanomaterial-based composites. Separating nanoparticles from composite matrices is often difficult and may not be achievable with complete efficiency [20]. Consequently, a top-down approach to sustainability that focuses on waste management, and strong product design is essential. By enhancing product

durability and extending service life, the environmental impact associated with the disposal of nanoparticle-reinforced composites can be reduced. Correia et al. explored this concept during their investigation of eco-friendly surfboards fabricated from bio-based, renewable materials [21]. The authors discovered that the mechanical performance of a surfboard made entirely from cork and wood-based materials did not meet industry standards and would result in needing to replace the surfboard more often, increasing overall product waste. Instead, the authors concluded that a hybrid approach of using fibre glass skin to reinforce the cork board resulted in a more robust design, creating a product with a longer lifetime. Using hybrid bio-based materials like SiNRs compared to entirely bio-based fillers may provide the optimum balance between mechanical properties and reduced environmental impact.

3. Materials and Methods

3.1. Materials

PET was obtained from cleaned Diet Coke bottles. SiNRs were supplied by Applied Quantum Materials (Edmonton, AB, CA) and were approximately 38 nm in diameter and 500 – 1000 nm in length. 99% trifluoroacetic acid (TFA) was purchased from Sigma-Aldrich (Edmonton, AB, CA). Rectangular substrates (25.6 × 77 mm) for spin coating were cut from a 1/16" Teflon sheet purchased from McMaster-Carr (Elmhurst, IL, USA). 5 mL Norm-Ject Luer Lock Solo syringes were purchased from Fischer Scientific (Waltham, MA, USA).

3.2. Preparation of SiNR-PET Solutions & Spin Coating Thin Films

SiNR and PET solutions of 20 wt% (solids in solvent) were prepared, with varied SiNR loadings to obtain compositions ranging from 0 to 5 wt% SiNR. SiNRs were first dispersed in TFA by sonication for 5 min, followed by magnetic stirring for an additional 2 hours. Small pieces of PET were cut from the Diet Coke bottle and added to the solutions. The mixtures were left to stir until the PET was fully dissolved. A Laurell Technologies Corporation spin coater (model WS-400B-6NPP/Lite) was used to make the composite films. The Teflon slides were cleaned with ethanol, and 2 mL of solution was dispensed on the surface, which was then spun over two cycles at 200 rpm and 2200 rpm/s acceleration for 1 minute before removing from the spin coater and left to air dry.

3.3. Tensile Testing

The films were completely dried before being carefully peeled away from the Teflon slides and cut into 10 mm × 77 mm strips for tensile testing. The average thickness of the films varied between 0.030 – 0.072 mm. A 1 kN MTS Insight load cell system was used for tensile testing. Following ASTM D882 for tensile testing of thin polymeric films, a gauge length of 27 mm and testing rate of 270 mm/min was chosen. Each composition was tested with a sample size of $n = 5$. Samples were tested to failure, as detected by the testing software. Elastic modulus was calculated by measuring the slope of the initial linear section of the engineering stress-strain curve. The yield strength (YS) was determined using the 0.2% offset method, defined as the intersection of a line parallel to the initial elastic modulus and offset by 0.2% strain with the engineering stress–strain

curve. Ultimate tensile strength (UTS) was the maximum stress a sample experienced during testing. All tests were measured at ambient conditions.

3.4. Microscopy

Scanning electron microscopy (SEM) images were acquired using a Hitachi S-4800 field-emission SEM (FESEM). Small sections of the composite films were mounted onto SEM stubs using carbon tape and sputter-coated with around 12 nm of gold prior to imaging. Transmission electron microscopy (TEM) images were acquired of the supplied SiNRs using a JEOL JEM-ARM200cF scanning transmission electron microscope (STEM). SiNRs were dispersed in ethanol and drop-cast onto TEM grids for imaging.

3.5. Thermogravimetric Analysis

Thermogravimetric analysis (TGA) of the neat PET and 5 wt% SiNR-PET films was conducted using a SDT Q600 TGA/DSC system. Small strips weighing around 5 mg of each sample were placed in an aluminum pan and heated from 20 to 750°C under nitrogen purge at a heating rate of 10°C/min.

4. Results and Discussion

The reinforcing effect a filler material imparts is highly susceptible to its compatibility with the polymer matrix. The surface of unmodified SiNRs are rich in –OH groups, which enhances compatibly with polymers that share polar functional groups, such as polyvinyl alcohol (PVA), nylon, polyvinyl chloride (PVC), polyurethanes (PUs), and PET [22]. The SiNRs interact with the polymer chains in the matrix through hydrogen bonding, creating a strong reinforcing network, as shown in Figure 1.

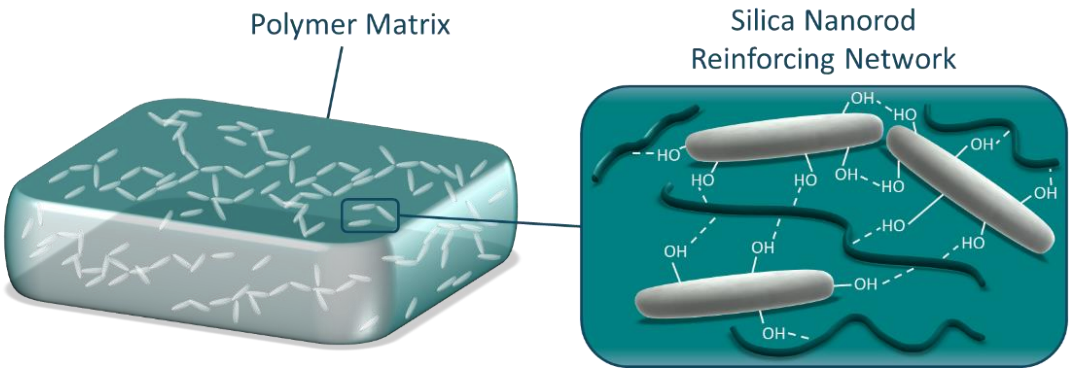

Figure 1. Schematic of the reinforcing effect of silica nanorods through hydrogen bonding.

The morphology and structure of the SiNRs used to make the composite films is evaluated using TEM, as shown in Figure 2. The TEM image shows long tubular strands with silica formed as beads around the surface of the CNCs. From the image, the width of the particles were measured as 21 nm and the lengths exceed 100 nm depending on the size of the individual CNC particle. The high surface area and bumpy texture due to the amorphous structure of the silica coating can promote stronger bonding forces between the SiNRs and polymer matrix.

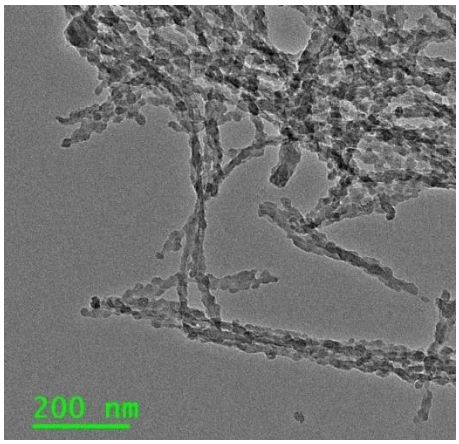

Figure 2. TEM image of SiNRs.

SEM was used to visualize the topography of the composite films. Since the loading of the SiNRs was low, individual nanoparticles were not easily visible during imaging. However, there was a noticeable influence from the nanoparticles on the surface roughness of the films. At low loadings (Figures 3a and 3b), the surface appears smooth with some small ridges. At higher loadings (Figures 3c and 3d), large ridges and bumps appear, potentially due to larger aggregates of the SiNRs, or the influence of the nanoparticles on the crystallization of the polymer when spin coating. This change in surface roughness is expected with increasing filler loading. Future tests using energy-dispersive x-ray spectroscopy (EDX) could aid in distinguishing the dispersibility efficiency of the SiNRs within the films.

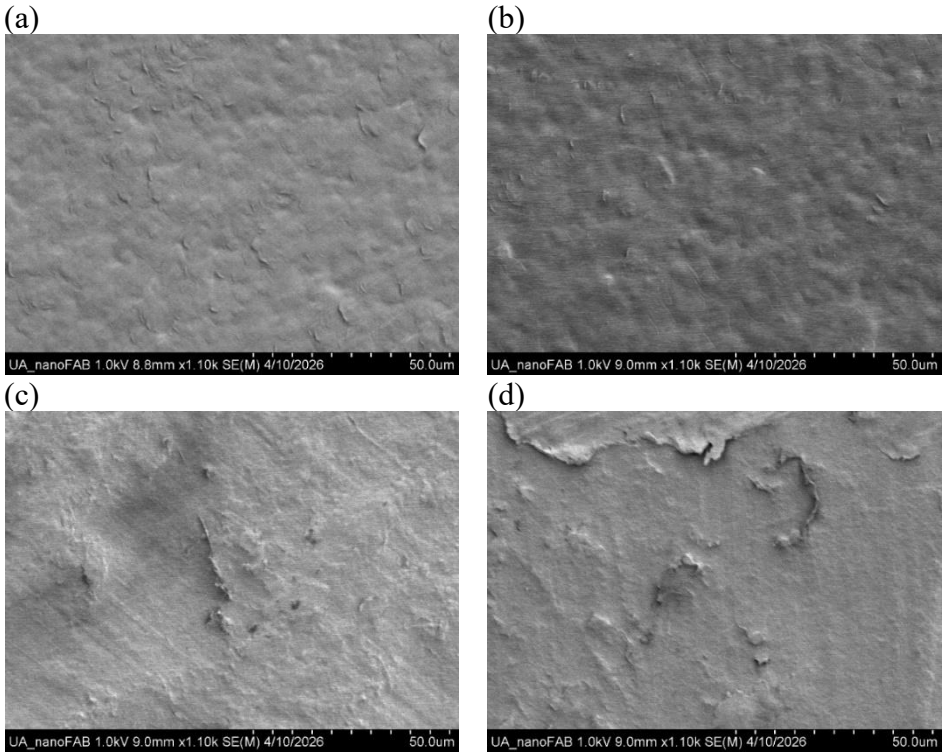

Figure 3. SEM images of SiNR-PET composites films (a) 0 wt% SiNR, (b) 1 wt% SiNR, (c) 2.5 wt% SiNR, and (d) 5 wt% SiNR.

Tensile tests were performed to evaluate the mechanical properties of the composite films. The elastic modulus increased by 72.8% (1.7×) between the neat PET and the 5 wt% SiNR films, as shown in Figure 4 and Table 1. These results demonstrate a clear reinforcing effect from the nanorods with increasing loading. Overall, UTS showed no statistically significant changes relative to neat PET within experimental error; however, the YS increased by 25.5% at a 5 wt% loading. The elongation followed a predicted trend of decreasing with increasing SiNR loading.

The significant improvement in stiffness and increase in yield strength suggests effective load transfer between the SiNRs and PET matrix. In polymer composites, decreased YS and UTS values typically indicate inefficient stress transfer within the matrix – exacerbated by incompatible fillers or agglomeration of filler particles – and increased plastic deformation [23, 24, 25]. The retention of UTS and increased YS suggests that the incorporation of the nanoparticles did not adversely affect interfacial adhesion within the matrix, which can be a common issue for bio-based fillers due to a mismatch in filler-matrix interactions.

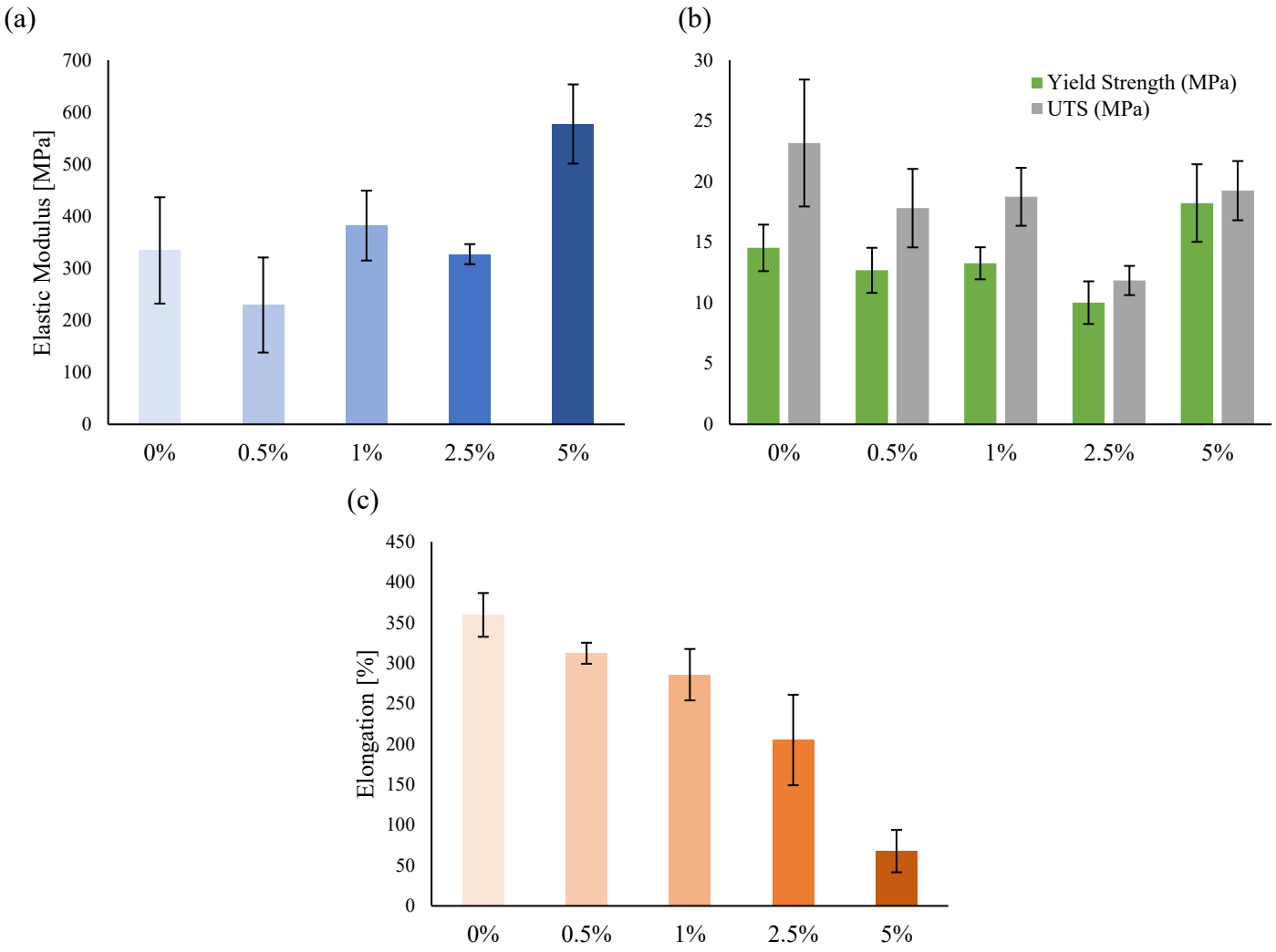

Figure 4. Mechanical test results of SiNR-PET composite films. (a) Elastic modulus (MPa), (b) combined yield strength (MPa) and UTS (MPa), and (c) elongation at break (%).

Table 1. Mechanical test results for SiNR-PET composite films.

SiNR content in PET (wt%)	Elastic Modulus (MPa)	YS (MPa)	UTS (MPa)	Elongation (%)
0	334 ± 102	14.5 ± 1.9	23.2 ± 5.2	360 ± 27
0.5	229 ± 91	12.7 ± 1.8	17.8 ± 3.2	312 ± 13
1	382 ± 67	13.3 ± 1.3	18.7 ± 2.4	286 ± 32
2.5	327 ± 19	10.0 ± 1.8	11.8 ± 1.2	205 ± 56
5	577 ± 76	18.2 ± 3.2	19.2 ± 2.4	68 ± 26

A key weakness of CNCs as nanofillers is their limited thermal stability and susceptibility to degradation during melt-mixing processing. The TGA results presented in Figure 5 show a representative TGA and DTG curve from three replicate tests for neat PET and 5 wt% loading SiNR-PET. The onset degradation temperature (T_o) and remaining mass/char shown in Figure 5a were largely unchanged. At approximately 200°C there is a small peak in the 5 wt% loading, which could be due to entrapped water within the SiNRs or residual unreacted TEOS. The mass loss mechanisms for the two compositions remained nearly identical, as can be seen in Figure 5b, which indicates that the SiNRs did not significantly change the degradation rate of the PET. Therefore, the incorporation of SiNRs into the PET matrix left the polymer's overall degradation behaviour largely unaffected.

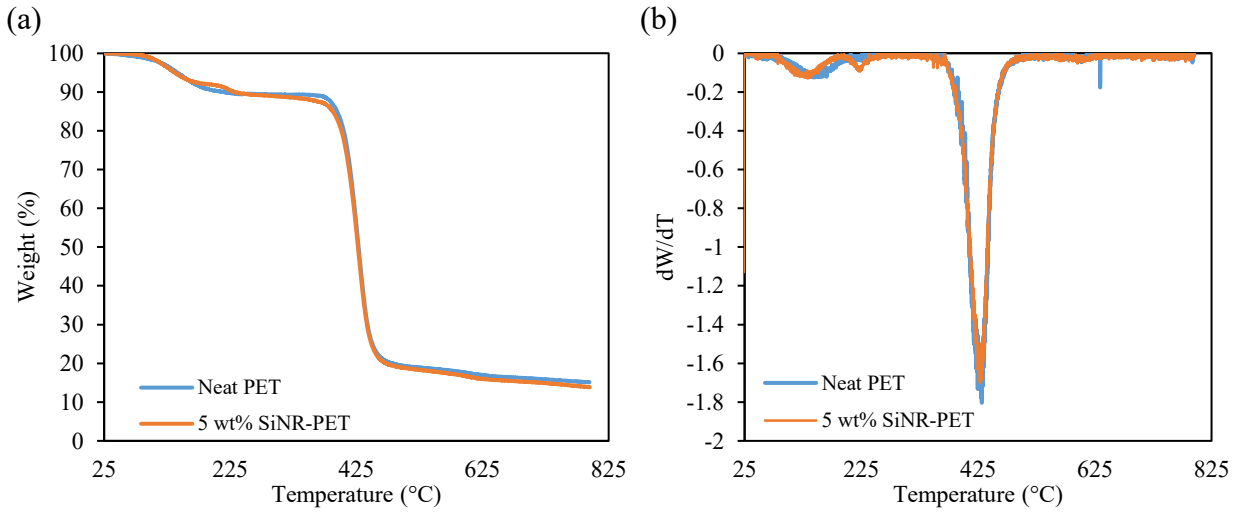

Figure 5. TGA (a) and DTG (b) results for neat PET and 5 wt% SiNR-PET.

5. Conclusion

SiNRs were evaluated as a sustainable hybrid filler material in PET thin-film composites, with the objective of assessing their influence on both mechanical performance and thermal stability. Loadings of up to 5 wt% SiNRs were evaluated for their mechanical performance using tensile testing, and their thermal stability was assessed using TGA. TEM and SEM were used to observe the morphology of the SiNRs and their interaction with the matrix material. The results showed an increase in elastic modulus of over 73% and a YS improvement exceeding 25% at 5 wt% loading of SiNRs. The incorporation of the SiNRs did not result in a significant change in the thermal stability of PET. Despite the limited understanding of the filler-matrix interactions from the SEM results, the increased YS and unchanged UTS values may indicate good interfacial adhesion

between the SiNRs and PET matrix. Additional characterization is needed to verify this assumption. However, the findings suggest that SiNRs are compatible with PET and show potential as a high-performance hybrid reinforcing filler.

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