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Preparation and Characterization of Modified Graphene-Doped Multi-walled Carbon Nanotube/Polypropylene Nanocomposite Films

Modifiye Grafen Dop Edilmiş Çok Duvarlı Karbon Nanotüp Katkılı Polipropilen Nanokompozit Film Üretimi ve Karakterizasyonu

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Abstract

In this study, polypropylene films were developed by incorporating both unmodified and chemically modified (oxidized and subsequently silanized) 32 wt% graphene-doped multi-walled carbon nanotubes (GR-doped MWCNTs) at three loading levels. In order to provide a homogeneous mixture of PP powder and the nanofiller, a non-traditional coagulation method was carried out. This method achieved the synergistic combination of modification-dispersion processes, which has not been conducted on GR-doped MWCNT-reinforced PP matrix composite before. The resulting blends were then hot-pressed under controlled temperature and pressure to produce nanocomposite films. The mechanical properties of modified and unmodified GR-doped MWCNT/polypropylene films were investigated via tensile testing. Subsequent thermal and morphological analyses were performed on the modified composites and neat polypropylene, since the elastic modulus of the modified films was generally higher than both neat polypropylene and unmodified GR-doped MWCNT-reinforced films. The fundamental thermal characteristics were determined by performing DSC (differential scanning calorimetry), TGA (thermogravimetric analysis), and DTA (differential thermal analysis) measurements. Both nanoparticle distribution and film surface morphology were evaluated via SEM (scanning electron microscopy). Based on thermal analysis, it was inferred that the presence of nanofiller did not exhibit any particular effects in terms of thermal characteristics (i.e., melting temperature or thermal degradation), which indicated that the PP sustained its thermal stability during the production stage. The results demonstrate that incorporating 3-aminopropyltriethoxysilane (APTES)-modified GR-doped MWCNTs into polypropylene enhances the elastic modulus of the composite films relative to neat polypropylene. These improved properties make the produced composite films suitable for use in lightweight automotive interior components, packaging films, and electronic casing applications.

Keywords: Graphene-doped multi-walled carbon nanotubes, nanocomposite polymer films, silane modification.

Öz

Bu çalışmada, polipropilen filmler; modifiye edilmemiş ve kimyasal olarak modifiye edilmiş (oksidasyon ve ardından silanizasyon gerçekleştirilmiş) 32 wt% grafen katkılı çok duvarlı karbon nanotüplerin (GR-doped MWCNTs) üç farklı yükleme oranında polimere ilave edilmesiyle geliştirilmiştir. PP tozu ve nano dolgu maddesinin homojen karışımını sağlamak için konvansiyonel olmayan bir metot olarak bilinen koagülasyon tekniği kullanılmıştır. Bu yöntem, daha önce GR-doped MWCNT takviyeli PP matris kompozite hiç uygulanmamış olan modifikasyon-dispersiyon işlemlerinin sinerjik bir kombinasyonunu sağlamıştır. Elde edilen karışımlar, kontrollü sıcaklık ve basınç altında sıcak pres tekniği kullanılarak nanokompozit film formunda üretilmiştir. Modifiye edilmiş ve edilmemiş GR-doped MWCNT/polipropilen filmlerin mekanik özellikleri çekme testi ile incelenmiştir. Modifiye filmlerin elastik

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modül değerleri, hem saf polipropilen hem de modifiye edilmemiş GR-doped MWCNT ile takviye edilmiş filmlerden genel olarak daha yüksek bulunmuştur, bu nedenle sonraki termal ve morfolojik analizler yalnızca modifiye kompozitler ve saf polipropilen üzerinde gerçekleştirilmiştir. Temel termal özellikler, DSC (diferansiyel taramalı kalorimetri), TGA (termogravimetrik analiz) ve DTA (diferansiyel termal analiz) ölçümleri yapılarak belirlenmiştir. Hem nanopartikül dağılımı hem de film yüzeylerinin morfolojisi SEM (taramalı elektron mikroskobu) ile değerlendirilmiştir. Termal analize dayanarak, nano dolgu maddesinin varlığının termal özellikler (erime sıcaklığı veya termal bozunma) açısından herhangi bir özel etki göstermediği, bunun da PP'nin üretim aşamasında termal kararlılığını koruduğu sonucuna varılmıştır. Elde edilen sonuçlar, 3-aminopropiltrietoksisilan (APTES) ile modifiye edilmiş GR-katkılı MWCNT'lerin polipropilene doplanarak üretilen kompozit filmlerin saf polipropilene göre elastik modülünü artırdığını göstermektedir. Geliştirilen bu özellikler, elde edilen kompozit filmin hafif otomotiv iç bileşenlerinde, paketleme filmlerinde ve elektronik gövde elemanlarında kullanıma uygun hale gelmesini sağlamaktadır.

Anahtar Kelimeler: Grafen dop edilmiş karbonnanotüp, nanokompozit polimer film, silanizasyon.

1. Introduction

Many thermoplastic polymers are commonly utilized in various applications, and they are exposed to different modifications to improve their performance. For instance, polypropylene is a well-known thermoplastic, and its isotactic semicrystalline form (i-PP) is mainly employed in diverse products. This form exhibits reliable thermal and mechanical behavior, along with acceptable physical properties (Karian, 2003), and is also relatively inexpensive. It is commonly used in many industries, including packaging and automotive. In addition to its resistance to gases and water, superior fatigue resistance, as well as improved impact strength and chemical stability, make PP appropriate for utilization in many industrial cases. It also finds application in areas such as textiles, orthopedic implants, electrical components, and various medical devices (Campo 2008; Bose & Bandyopadhyay 2017; Guarino & Ambrosio (eds.) 2018; Greene 2021; Sin & Tueen 2023).

However, neat PP shows poor fracture toughness and is highly brittle; it performs poorly in applications that require high impact resistance (Shirvanimoghaddam et al., 2021). Its high thermal expansion coefficient and oxidation sensitivity also reduce its performance in environmental conditions. To eliminate those limitations, micron- or nano-sized additives (such as talc, glass fiber, carbon nanotubes, and graphene) are consolidated with a PP matrix. The industrial (e.g., automotive, machinery, appliances, and construction) needs, such as high strength, rigidity, and wear resistance, can be promoted with proper filler(s) (Yang et al., 2008; Luo et al., 2018; Wang et al., 2020; Tsioptsias et al., 2024). Additionally, the heat resistance is enhanced by introducing glass-fiber or mineral additives having high heat-distortion temperatures (Altay et al., 2021; Caban, 2022). Among various nanoscale fillers, carbon nanotubes (CNTs) and graphene (GR) are of considerable interest. When incorporated into PP, they can improve the modulus and toughness while enhancing thermal and electrical conductivity (Imran et al., 2018; Hussein et al., 2024). However, their smooth, chemically inert surfaces, high specific surface area, and strong π–π van der Waals interactions often lead to agglomeration within the polymer matrix (Atif & Inam, 2016). The phenomenon increases with higher filler loadings, so it is often reported at low concentrations in published studies (Zeinedini et al., 2025). To compensate for dispersion, hybrid GR-doped CNT architectures have been engineered into two kinds. The 2D GR sheets provide large, planar surfaces that support a more uniform CNT distribution. On the other hand, GR-wrapped CNTs exhibit smooth, protective interactions, resulting in synergistic improvements in mechanical and/or thermal properties (Liu et al., 2025).

Despite extensive research on CNTs and graphene regarding their mechanical and thermal performance, studies on graphene-doped CNT reinforcement remain limited. Therefore, the nanofiller's surface chemistry strongly influences the resulting composite performance. Although many efforts to improve matrix–filler adhesion, including covalent, non-covalent, and elemental-doping strategies (Yu et al., 2020; Gao et al., 2023), interfacial bonding continues to be a limiting factor in polymer nanocomposites. Among these methods, silane modification remains widely regarded as a practical and effective approach for carbon-based nanoparticles. First, nanoparticle surfaces are oxidized (acid or $KMnO_4$ treatment) to introduce –OH/–COOH groups; then organosilanes are grafted to form strong interfacial bonds, resulting in improved load transfer, dispersion uniformity, and both mechanical and thermal performance (Salihu et al., 2011).

3-aminopropyltriethoxysilane (APTES) was selected because of its strong affinity for defect sites on oxidized MWCNT surfaces arising from its amino functional groups (Kathi & Rhee, 2008; Lavorgna et al., 2013). Its flexible aliphatic chain helps reduce agglomeration and improves

compatibility with non-polar polymers such as polypropylene (Lee et al., 2010; Omar et al., 2014). As a result, the alkyl chain interacts with polypropylene through van der Waals interactions, thereby enhancing the dispersion of MWCNTs within the PP matrix.

In order to gain the optimum properties of the nanofiller, the oxidation followed by silane treatment of GR-doped MWCNTs was performed. During the oxidation step, –OH and –COOH groups were introduced onto the surface of the GR-doped MWCNTs, creating defect-rich regions (Datsyuk et al. 2008, Sahebian et al. 2015). This was followed by silanization with APTES, which formed Si–O–C linkages on the surface (Kathi & Rhee, 2008; Lavorgna et al., 2013). The resulting modified nanofiller allows stronger interactions with the nonpolar PP matrix and improves dispersion. This structural configuration contributes to the observed increase in elastic modulus and, consistent with previous reports, aminosilane-functionalized CNTs are known to enhance interfacial bonding, stress transfer, and dispersion quality in polypropylene nanocomposites (Ma et al., 2010; Lavorgna et al., 2013; Pang et al., 2013).

The novelty of this study lies in the combined use of chemical silanization and coagulation-assisted dispersion for a 32 wt% graphene-doped MWCNT hybrid filler, which further improves filler distribution.

2. Materials and Methods

In the experimental study, neat polypropylene films were first produced as a reference. Three concentrations (0.05, 0.025, and 0.01 wt%) of GR-doped MWCNT powder were then incorporated into the polypropylene matrix to fabricate nanocomposite films using the coagulation technique, as reported in the literature (Novikov et al., 2022) (Figure 1). Additionally, the GR-doped MWCNTs were surface-functionalized via silane modification. Using modified and unmodified GR-doped MWCNTs, three sets of polypropylene composite films (0.05, 0.025, and 0.01 wt% filler loadings) were prepared.

2.1. Material

The GR-doped MWCNT powders employed in this study were sourced from Nanografi, toluene was obtained from Isolab, and the i-PP was provided by Petkim.

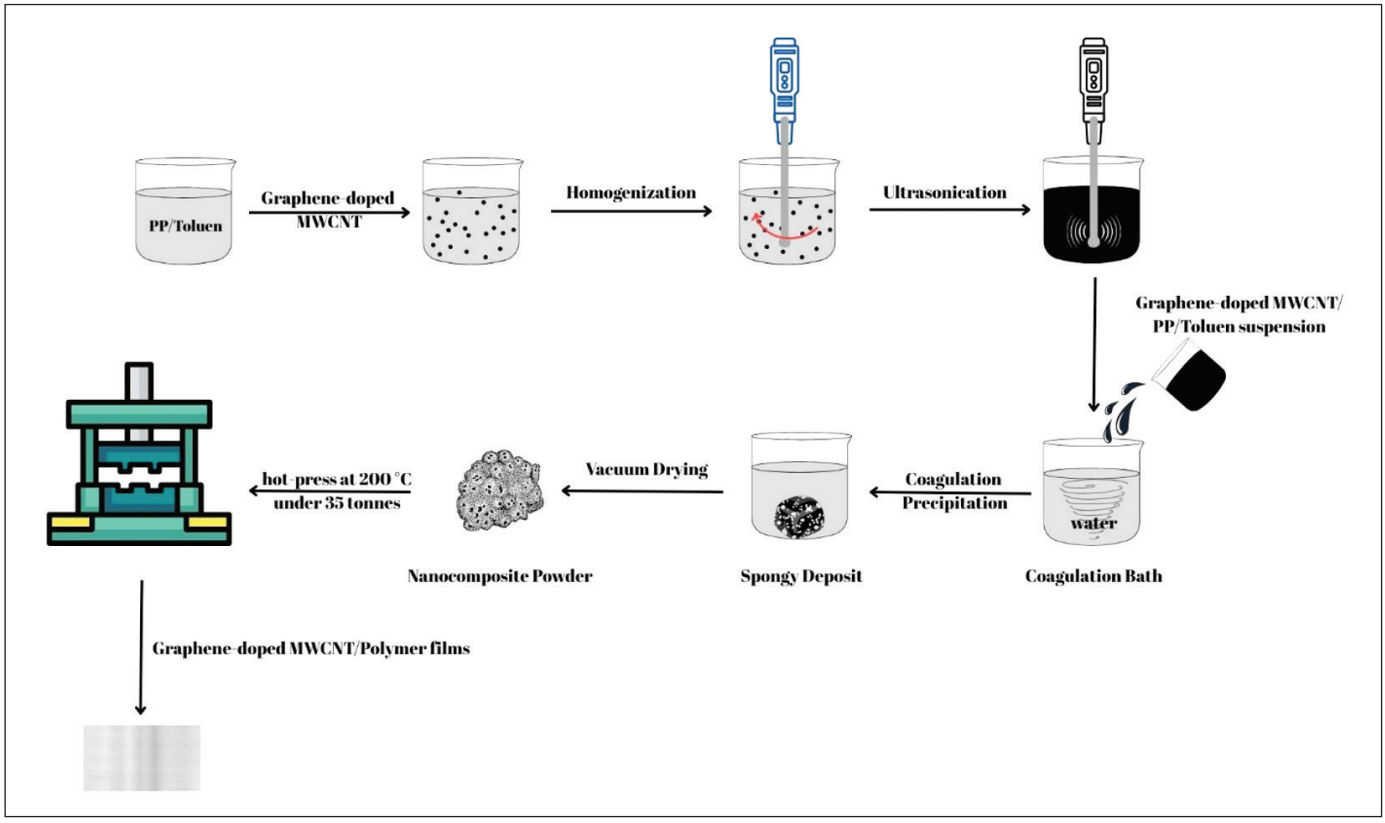

Figure 1. Production of modified and unmodified GR-doped MWCNT/PP nano composite films via the coagulation method.

2.2. Method

The Raman measurements were carried out in backscattering geometry with a Renishaw spectrometer and a confocal microscope, using a 532 nm excitation wavelength. A Shimadzu Autograph A65-X universal testing machine was used to perform the tensile tests. For this test, rectangular strip specimens (10 mm width x 140 mm length) with an average thickness of 60-80 μm were prepared according to ASTM D882. The gauge length of 50 mm was determined for all measurements. During testing, a prestress of 0.1 MPa was used, and the crosshead speed was maintained at 1 mm min^{-1}. Differential scanning calorimetry (DSC) was carried out on a TA Instruments DSC 250 to determine the melting temperature, crystallinity, and melting enthalpy parameters. Each sample was subjected to a heat–cool–heat cycle from −80 to 300 °C, using a heating/cooling rate of 20 °C min^{-1}. Non-isothermal cooling thermograms were recorded separately at the same cooling rate (20 °C min^{-1}). Thermogravimetric analysis (TGA) was performed using a Tetra Az TG analyzer under a N_2 atmosphere at *a 5 °C min^{-1} heating ra*te over 25–600 °C to evaluate thermal stability and mass-loss profiles. Surface morphology and nanoparticle dispersion were analyzed by scanning electron microscopy (LEO-EVO 40). SEM samples were cut into 5 x 5 mm^2 sections and sputter-coated prior to imaging using a BAL-TEC SCD 050.

2.2.1. Oxidation of GR-Doped MWCNT

GR-doped MWCNTs were subjected to an oxidation process to introduce oxygen-containing functional groups and increase surface reactivity using sulfuric and nitric acids. For this procedure, 0.0692 g of GR-doped MWCNT was poured into a mixture of 60 mL H_2SO_4 and 20 mL HNO_3. The mixture was heated in an oil bath and stirred with a magnetic stirrer at 50 °C. Before filtration and washing out with distilled water, the stirring process was continued at this constant temperature for 20 hours.

This washing process was repeated until the mixture reached pH 5. The resulting solid was vacuum-filtered and then dried in an oven for 4–5 days. The oxidation conditions were selected based on widely used acid functionalization protocols for carbon-based nanofillers (Vast et al. 2004; Chiu et al. 2008).

2.2.2. Silane Modification of GR-Doped MWCNT

Silane modification was applied to GR-doped MWCNTs to enhance the interfacial interaction between the nanofiller and the inherently non-polar polypropylene matrix. This modification is expected to improve the dispersion quality and, thereby, improve the mechanical and thermal properties of the nanocomposites. For silanization, 0.0692 g of oxidized GR-doped MWCNT and 2% (v/v)

3-aminopropyltriethoxysilane was dispersed in 300 mL of an ethanol-water solution (95:5). The dispersion was stirred with a magnetic stirrer at 70 °C for 4 hours. Subsequently, the solid was filtered, washed with distilled water and acetone, and then dried in an oven at 80 °C for 12 hours. Silane grafting parameters were adapted from established aminosilane modification methods for carbon nanomaterials to enhance interfacial compatibility (Lee et al., 2010; Mashhadzadeh et al., 2017).

2.2.3.Production of Modified and Unmodified GR-Doped MWCNT/PP Nano Composite Films:

2.2.3.1. The Coagulation Process

PP powder was dissolved in 200 mL of toluene at 25 °C under magnetic stirring for 1 h. The GR-doped MWCNTs were added to the PP solution and stirred magnetically for 1 h. The resulting suspension was subjected to high-speed homogenization at 9000 rpm for 15 min. Dual mixing, a combined process of ultrasonic treatment and magnetic stirring, was then applied for 1 h to improve dispersion. To promote coagulation, the well-mixed suspension was poured into 800 mL of deionized water and vortex-agitated. Stirring continued for 1 h to complete GR-doped MWCNT/PP precipitation. The coagulated GR-doped MWCNT/PP was filtered and transferred to an oven at 80 °C for 5–7 days, until fully dried, yielding a fine powder. The coagulation-assisted precipitation method was implemented following previously reported nanocomposite preparation routes (Mazov et al., 2011; Novikov et al., 2022).

2.2.3.2. Film Formation by Hot-Pressing

Neat PP and the dried composite powder (containing 0.01, 0.025, or 0.05 wt% unmodified GR-doped MWCNT and modified GR-doped MWCNT) was placed between aluminum-foil-lined metal plates. All films were fabricated in a hot-press at 200 °C under 3-4 MPa for 20 minutes (including heating and dwell time). These processing conditions are consistent with reported hot-press parameters used for PP-based composites in the literature (Merter 2009; Deng et al., 2010).

3. Results and Discussion

3.1. Raman Spectroscopy

Raman spectroscopy was used to evaluate how oxidation and subsequent silanization altered the structural order and

defect density of the GR-doped MWCNTs. Spectra were collected from two regions (Region 1 and Region 2) for neat, oxidized, and silane-modified samples. The peak positions and D/G ratios were compared (Table 1). The D band (~1340 cm^{-1}), associated with structural disorder and defect density in the sp^2 carbon network (Piao et al., 2021), is clearly observed in Figure 2. After oxidation, both the D-band intensity and the D/G ratio increased. Following silanization, the D-band intensity decreased relative to the oxidized GR-doped MWCNTs, but the D/G ratio remained slightly higher than that of the unmodified sample, confirming successful functionalization. The G band (~1570–1590 cm^{-1}), associated with in-plane vibrations of sp^2 carbon atoms, shifted to higher wavenumbers (from 1578 to 1590 cm^{-1}) after oxidation and slightly lower (~1575 cm^{-1}) after silanization, due to electron-withdrawing oxygen groups and electron-donating silane groups (Gopalakrishnan et al., 2012). Region 2 showed a higher D/G ratio (1.154) for the silane-modified sample compared to Region 1 (1.078), indicating more pronounced surface defects (Murray et al., 2021). The G' band (~2670–2700 cm^{-1}) exhibited a minor red shift after silanization, suggesting slight changes in graphitic stacking. The FWHM of the G band increased after oxidation and silanization in Region 1, while in Region 2 it narrowed, indicating local variations (Muzyka et al., 2018).

Table 1. Raman spectra data of GR-doped MWCNT, oxidized GR-doped MWCNT, and silane-modified GR-doped MWCNT (Regions 1 and 2).

Sample	Region	G (cm^{-1})	G FWHM	D (cm^{-1})	D FWHM	D/G	G′ (cm^{-1})	G′ FWHM
GR-doped MWCNT	R1	1578.80	156.16	1345.83	257.54	1.046	2684.72	549.03
	R2	1578.81	185.35	1343.56	222.49	1.042	2693.84	662.68
Oxidized GR-doped MWCNT	R1	1589.94	174.97	1344.56	241.35	1.070	2686.90	947.73
	R2	1590.84	177.65	1351.50	250.26	1.039	2686.54	973.55
Silane-modified GR-doped MWCNT	R1	1575.52	173.91	1340.16	208.21	1.078	2673.75	969.02
	R2	1575.52	143.97	1342.43	160.66	1.154	2664.59	1235.20

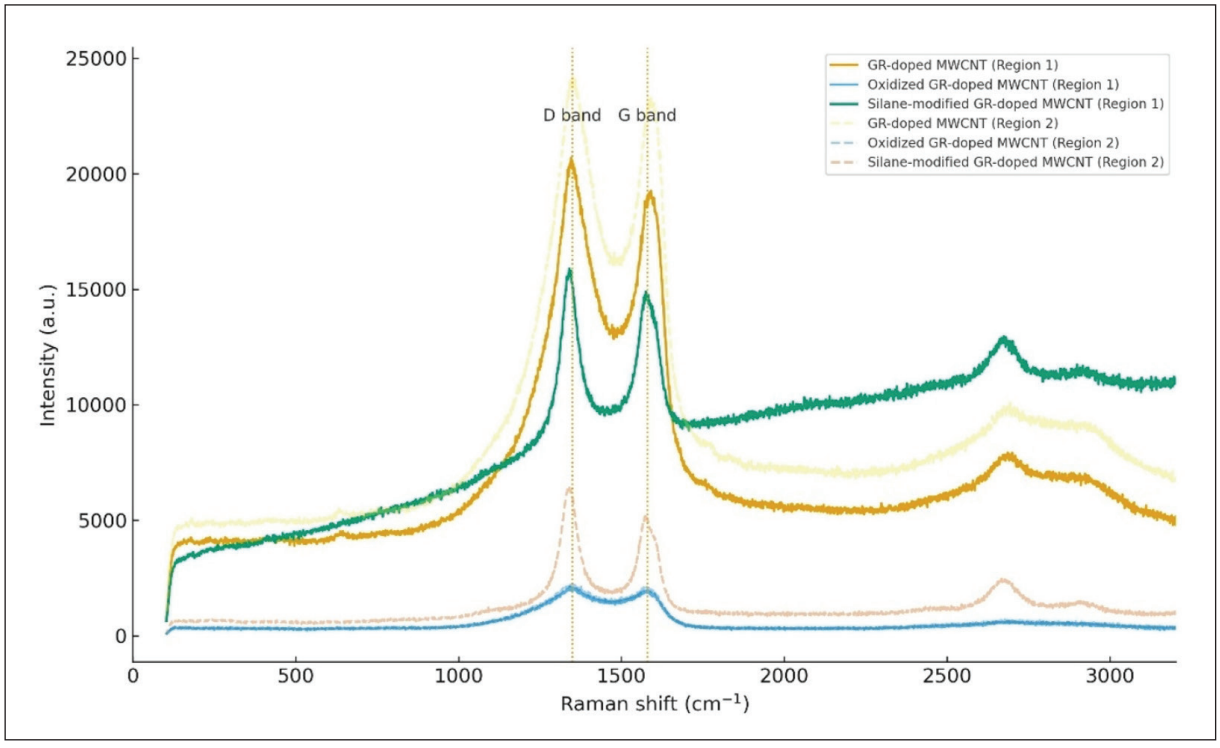

Figure 2. Raman spectra of GR-doped MWCNT, oxidized GR-doped MWCNT, and silane-modified GR-doped MWCNT (Regions 1 and 2).

3.2. Tensile Test Results

To evaluate the effect of silane modification on the mechanical behavior of GR-doped MWCNT/PP composites, tensile tests were performed at different filler loadings. Table 2 reports the elastic modulus and tensile strength values for composite materials, including different concentrations of modified and unmodified additives. As observed from the tensile test results, silane modification of GR-doped MWCNTs affected the stiffness and strength of composite films to varying degrees. According to Table 2, composites with silane-treated particles generally showed an improved elastic modulus at all filler amounts. The highest increase (9%) was observed in the composite film containing 0.01 wt% silane-modified nano-powder. However, the 0.05 wt% GR-doped MWCNT sample exhibited a higher modulus than neat PP.

The strength parameter of silane-treated composites also exhibited enhanced values. In particular, the specimens with 0.01 wt% silanized nanofiller showed a 48% improvement compared with the unmodified PP sample. A similar trend is also observed for the other composites with 0.025% and 0.05% filler concentrations. Those samples whose particles were exposed to modification exhibited higher strength values, albeit at lower percentages (see Table 2).

The effects of silane modification for each particle amount in composites are separately plotted as stress-strain curves in Figures from 3 to 5. When the unmodified 0.01 wt% GR-doped MWCNT was added to the PP matrix, the composites exhibited a more brittle characteristic, as indicated by a decrease in failure strain (see Figure 3). In contrast, silane modification changed that behavior and enhanced ductility with a maximum elongation at break (~0.06 mm/mm strain) as well as maximum strength (33.34 MPa UTS value). The silanization of 0.025 wt%. GR-doped MWCNTs led to slight improvements in terms of both stiffness and strength, as shown in Figure 4. Despite that progress, tensile strength was lower, and the elastic modulus was slightly higher than that of the pure matrix for this concentration. Unlike in the previous composite system, the ductility of neat PP was reduced by the incorporation of fillers, whether or not they were silanized. That sort of response was observed for the 0.05 wt%. filler introduced composite films as well (see Figure 5). However, chemical modification enhanced the plastic deformation capability of the particles and resulted in improved strength and strain values compared to unmodified additive-filled films.

Based on these findings, the optimal concentration of silane-modified GR-doped MWCNTs is determined as 0.01 wt%. Silane treatment improved mechanical properties by chemically linking GR-doped MWCNTs to other nanostructures and enhancing compatibility with the polymer matrix. This promoted more uniform stress transfer within the composite, enabling a larger portion of the applied force to be carried by the polymer, ultimately enhancing the

Table 2. Comparison of tensile samples of neat PP, silane-modified, and unmodified GR-doped MWCNT/PP films.

Sample / Characterization	Neat PP	0.01 wt % GR-doped MWCNT	0.025 wt % GR-doped MWCNT	0.05 wt % GR-doped MWCNT
Elastic Modulus (MPa)	1366.93 (±55.36)	1298.73 (±47.21)	1297.9 (±267.07)	1648.63 (±100.52)
σ Maximum Strength (MPa)	27.82 (±1.50)	22.62 (±1.52)	23.38 (±5.15)	27.44 (±1.44)
Sample / Characterization	**Neat PP**	**0.01 wt % silane-modified GR-doped MWCNT**	**0.025 wt % silane-modified GR-doped MWCNT**	**0.05 wt % silane-modified GR-doped MWCNT**
Elastic Modulus (MPa)	1366.93 (±55.36)	1518 (±180.59)	1407.23 (±78.42)	1485.97 (±75.52)
σ Maximum Strength (MPa)	27.82 (±1.50)	33.34 (±2.65)	26.22 (±2.22)	28.31 (±4.11)

material's strength. However, in the unmodified GR-doped MWCNT-reinforced composites at different concentrations, the elastic modulus was reduced except for 0.05 wt%, which was probably attributed to nanoparticle agglomeration in the polymer matrix (Bikiaris, 2010).

In the literature, comparable or higher improvements in elastic modulus (≈10–40%) are typically reported at CNT loadings of 0.1–3 wt% (Prashantha et al., 2009; Choi et al., 2009). In contrast, in this study, an ≈11% increase in elastic modulus was achieved at only 0.01 wt% silanized GR-doped MWCNT, highlighting the efficiency of the combined silanization and coagulation-assisted dispersion strategy.

3.3. Differential Scanning Colorimetry Analyses (DSC)

DSC analysis was performed to evaluate how silane-modified GR-doped MWCNTs influence the melting behavior and crystallization characteristics of polypropylene. From the peak areas of these curves (Figures 6 and 7), crystallization enthalpies were obtained, and the degree of crystallinity (Xc) of the composites was evaluated using the heat released during crystallization (ΔHc) according to the following equation:

$$X_c\ (\%) = \frac{\Delta Hc}{(1 - wt\%)\Delta Hm} x100$$

where **ΔHm** is the heat of fusion for 100% crystalline isotactic PP (ΔHm = 209 J/g), and **wt%** represents the weight fraction of MWCNTs in the composite (Mark, 1996).

As the GR-doped MWCNT content increased, a slight decrease (~2 °C) in the melting temperature (T_m) was observed, likely due to a reduction in crystallite size (Coppola et al. 2020). According to Table 3, the 0.01% silane-modified GR-doped MWCNT addition did not significantly alter crystallinity compared to neat PP, whereas the 0.025% addition slightly decreased it. However, at a 0.05% loading, crystallinity increased by approximately 3%. During the second heating cycle (i.e., after eliminating thermal history associated with processing), no significant differences in melting temperature were observed among the samples, with melting points around 161 ± 1 °C (Table 3), while the degree of crystallinity of the GR-doped MWCNT/PP nanocomposites increased by approximately 3% at a 0.05% loading compared to neat PP. The literature reports that MWCNTs can act as nucleating agents in the crystallization of PP with no significant change (Leelapornpisit

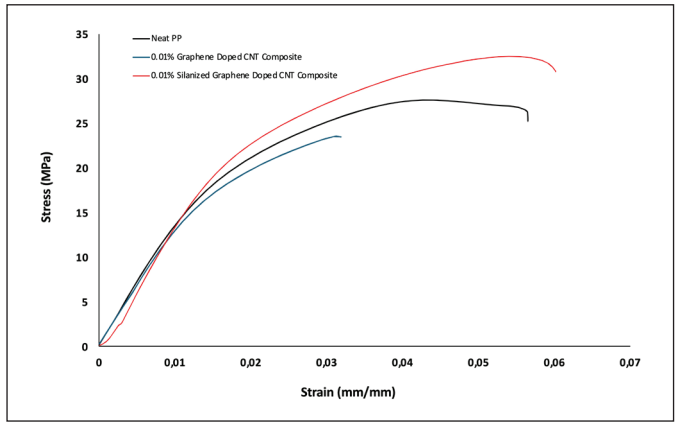

Figure 3. Representative stress-strain graphs of neat PP, modified, and unmodified 0.01 wt% GR-doped MWCNTs/PP films.

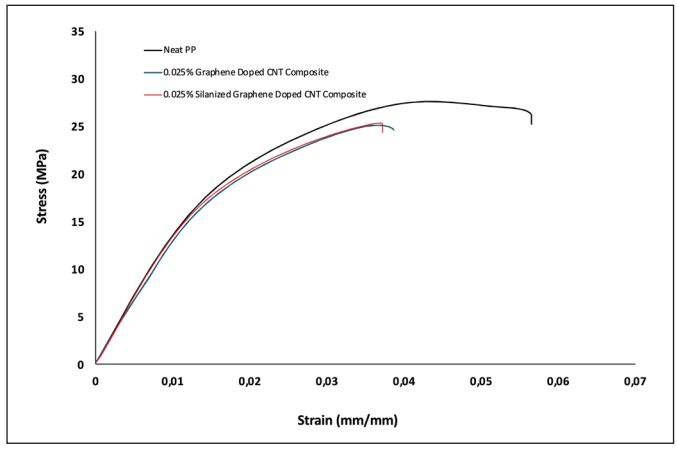

Figure 4. Representative stress-strain graphs of neat PP, modified, and unmodified 0.025 wt% GR-doped MWCNTs/PP films.

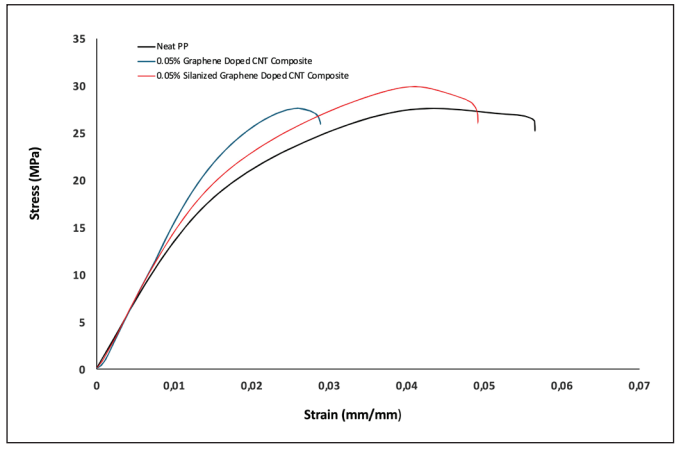

Figure 5. Representative stress-strain graphs of neat PP, modified, and unmodified 0.05 wt% GR-doped MWCNTs/PP films.

et al., 2005; Tabuani et al., 2007; Xu & Wang 2008), and PP/MWCNT nanocomposites commonly exhibit a pronounced nucleating effect leading to increased crystallinity (Bhuiyan et al., 2013). In contrast, in this study, the addition of 0.01 wt% silanized GR-doped MWCNTs did not produce a significant change in the DSC crystallinity of the PP matrix (Xc ≈ 46%). Therefore, the enhancement in elastic modulus can be attributed primarily to filler rigidity and interfacial interactions rather than to any crystallinity-related effects.

3.4. Thermogravimetric Analyses (TGA)

TGA analysis was performed to assess whether incorporating silane-modified GR-doped MWCNTs affects the thermal degradation behavior of polypropylene. According to TGA, the decomposition temperatures of the composite films ranged from 400 to 480 °C (Figure 8). It is evident from the thermograms that all polymer samples, both neat and silane-modified, GR-doped, and MWCNT-reinforced, underwent single-step thermal degradation. In terms of ther-

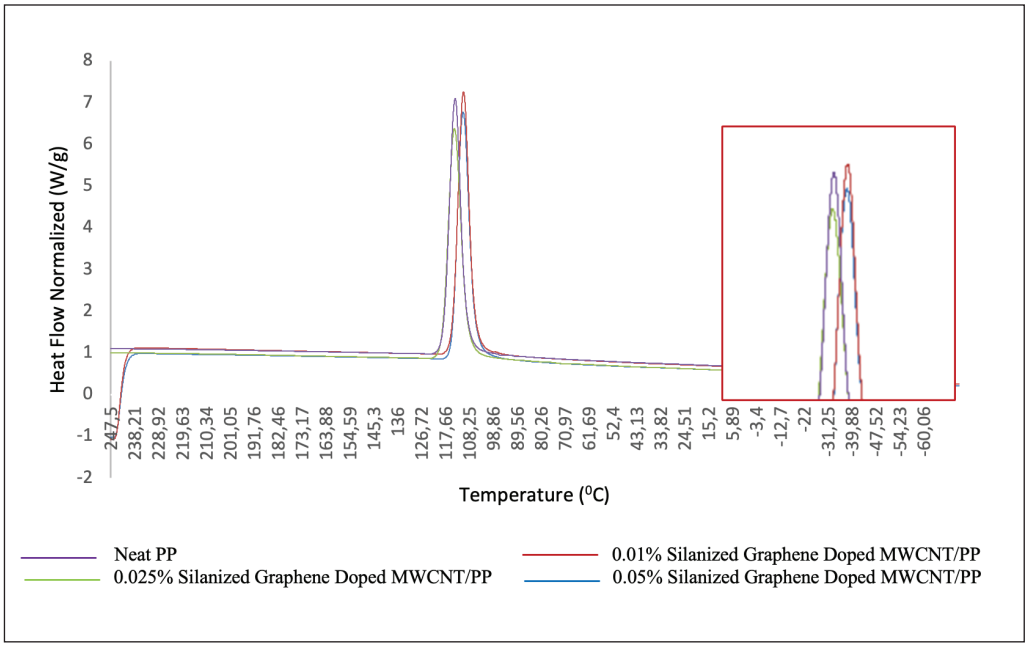

Figure 6. DSC cooling curves of neat PP and 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP film samples.

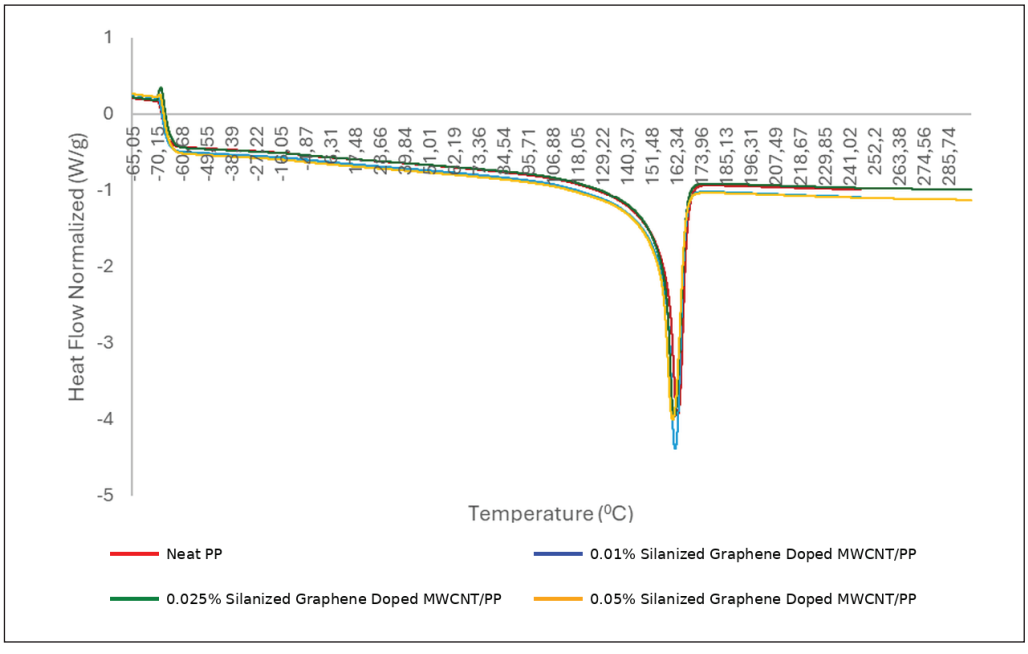

Figure 7. DSC second heating curves of neat PP and 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP film samples.

Table 3. DSC enthalpy (j/g), melting peak temperature (°C), and crystallization rate (%) data of neat PP and 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP film samples.

	Entalphy (Heating) J/g	T_m (Melting) Peak Temperature (Heating) ^{0}C	X_c (%)
Neat PP	95.169	162.69	45.54
0.01% silane-modified GR-doped MWCNT/PP	94.164	161.83	45.51
0.025% silane-modified GR-doped MWCNT/PP	89.551	161.49	43.95
0.05% silane-modified GR-doped MWCNT/PP	96.093	160.73	48.40

mal response, there was no significant difference between silane-modified GR-doped MWCNT/PP composite films and neat polypropylene, with their initial and final degradation temperatures also being nearly the same.

The mass loss values for neat PP and 0.05 and 0.025 wt% silane-modified GR-doped MWCNTs/PP were determined as 99%, 99.3%, and 99.5%, respectively. Nevertheless, the composite film with 0.01 wt% silane-modified GR-doped MWCNT showed a slightly lower mass loss of 97.3%, corresponding to a minor change compared to other specimens.

The onset degradation temperatures of neat PP and the nanocomposite were nearly identical (≈423–424 °C), indicating that such a low CNT loading does not significantly influence the initial decomposition stages. Similar behavior has been reported for PP/MWCNT systems in earlier studies, and at low filler loadings, CNTs cause only very small changes in the Tonset value (Ávila-Orta et al., 2013; Han et al., 2018; Naddeo et al., 2017). Similarly, the end of the main degradation step occurred at approximately 469 °C for both samples, falling within the typical TGA range (≈460–475 °C) reported for PP-based CNT nanocomposites (Han et al., 2018; Cabello-Alvarado et al., 2019).

The total mass loss decreased from 99% in neat PP to 97% in the nanocomposite containing 0.01 wt% MWCNT. This decrease is mainly due to CNTs remaining thermally stable as char under the applied conditions (Kashiwagi et al., 2002; Zhou et al., 2016). According to TGA data, the degradation profile remained similar to that of neat PP at a 0.01 wt% filler loading, with a small increase in the final residue, consistent with CNT stability.

Similarly, DTA data revealed no significant changes in the peak temperatures. The DTA curve exhibited two endo-

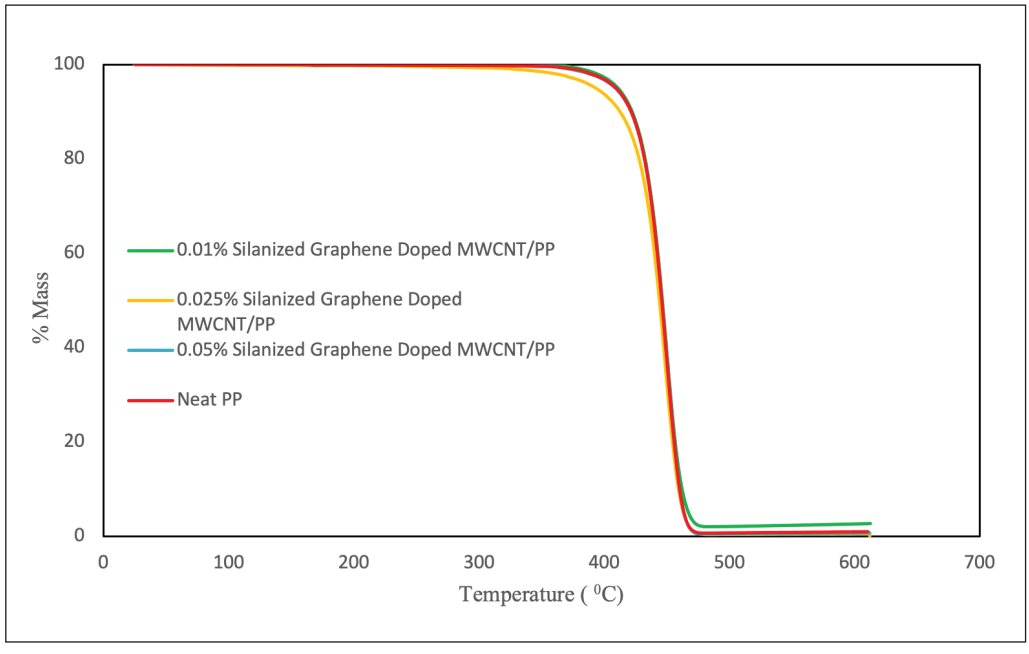

Figure 8. TGA curve of % weight loss for 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP nanocomposites.

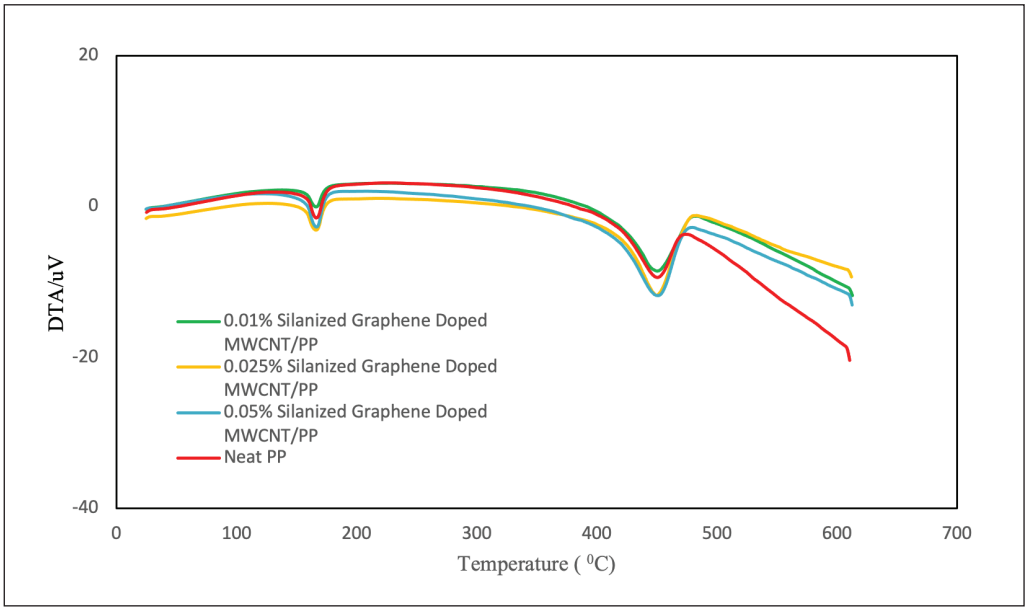

Figure 9. DTA curve of % differential thermal analysis for 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP nanocomposites.

thermic peaks: the first corresponds to the melting peak at 168 °C, and the second corresponds to the decomposition peak at 453 °C (Figure 9).

DTG provides information on the rate of maximum mass loss during decomposition. According to the DTG analysis, the decomposition onset temperature was 422.8 °C, with a peak decomposition temperature of 450.9 °C and an end temperature of 468.7 °C; these values were nearly identical across all derivatives (Figure 10). The highest mass loss rate was observed in the composite containing 0.05% silane-modified GR-doped MWCNT. In contrast, the degradation rate was similar in the neat PP and the sample with 0.025% silane-modified GR-doped MWCNT. The 0.01 wt% silanized GR-doped MWCNT/PP sample showed a slight increase in the DTG peak temperature compared to neat PP, indicating an improvement or at least preservation of thermal stability.

Most studies reporting significant increases in thermal degradation temperature (DTG peak temperature) (+10 to +35 °C) involve CNT loadings of 0.5–1 wt% or higher (Sahli et al., 2020; Kashiwagi et al., 2004). In contrast, studies employing lower CNT contents often report only small or inconsistent changes in DTG peak temperature. These findings indicate that very low CNT concentrations have only a limited influence on the degradation kinetics of the polymer, and that the effect becomes pronounced only as the filler loading increases. Overall, the DTG results corroborate the TGA findings and demonstrate that at 0.01 wt% silane-modified GR-doped MWCNT loading, the thermal degradation behavior of PP is largely maintained, with only small shifts in the peak degradation temperature and a slight modification of the degradation rate profile.

3.5. Scanning Electron Microscopy Results (SEM)

SEM analysis was used to evaluate the dispersion quality and surface distribution of the silane-modified GR-doped MWCNTs within the PP matrix. When examining SEM images of the surfaces of polypropylene films containing silane-modified fillers, it is observed that at the lowest loading level of 0.01 wt%, particles are well dispersed, with particle sizes around 200 nm (Figure 11). However, as the loading percentage increases, dispersibility decreases slightly, and fewer particles are observed on the surfaces. Additionally, due to agglomeration, the additive size increases to approximately 1 μm (Figure 12). The particles with a 0.05 wt.% concentration embedded in the PP matrix are shown in Figure 13.

As summarized in Table 4, the mechanical and thermal properties of neat PP and the 0.01 wt% silane-modified GR-doped MWCNT/PP nanocomposite are compared with typical improvements reported for CNT/PP systems in the literature.

4. Conclusion and Suggestions

In this study, the graphene-doped multi-walled carbon nanotubes (32 wt% GR-doped MWCNTs) hybrid powder

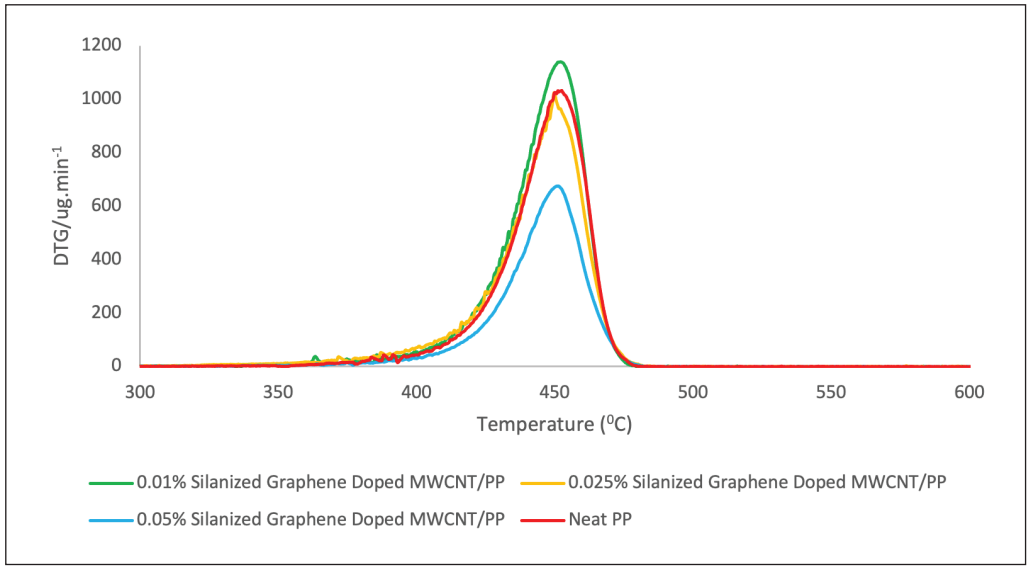

Figure 10. DTG curve of % differential thermal analysis for 0.01, 0.025, and 0.05 wt% silane-modified GR-doped MWCNT/PP nanocomposites.

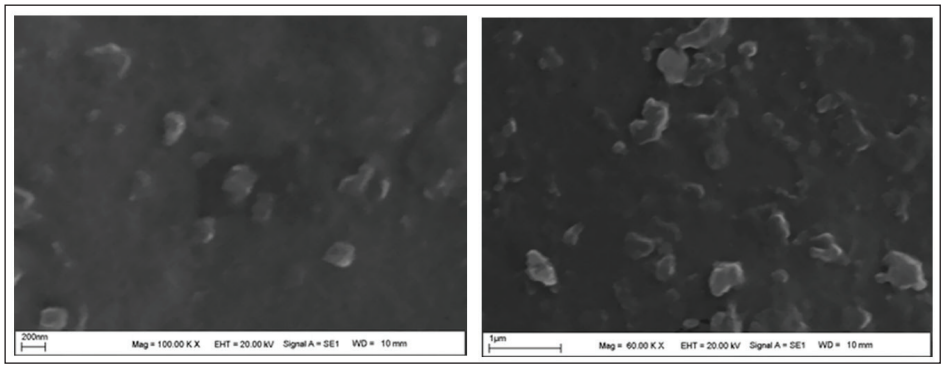

Figure 11. SEM image of the surface of a polypropylene film containing 0.01 wt% silane-modified GR-doped MWCNTs.

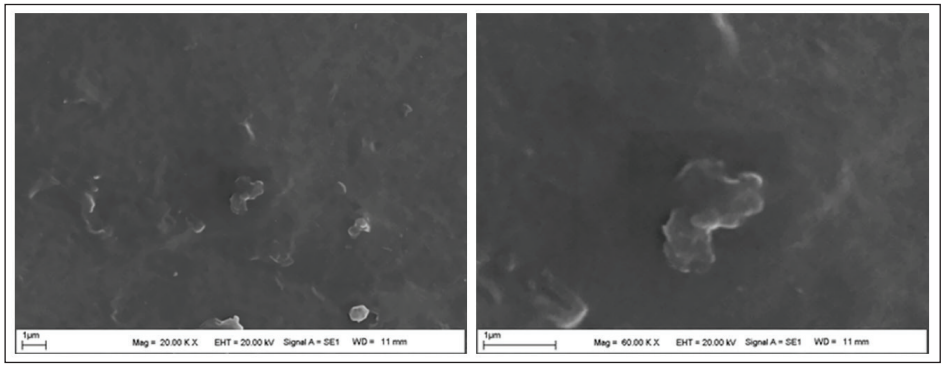

Figure 12. SEM image of the surface of a polypropylene film containing 0.025 wt% silane-modified GR-doped MWCNTs.

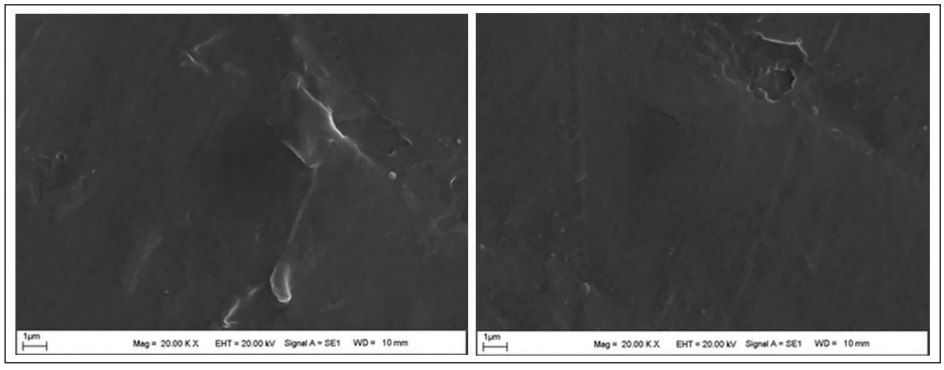

Figure 13. SEM image of the surface of a polypropylene film containing 0.05 wt% silane-modified GR-doped MWCNTs.

Table 4. Comparison of mechanical and thermal properties of neat PP and 0.01 wt% silane-modified GR-doped MWCNT/PP nanocomposite, alongside typical improvements reported in CNT/PP systems in the literature.

Property	Neat PP (This study)	0.01 wt% Silane-Modified GR-MWCNT/PP (This study)	Improvement (%)	Typical CNT/PP or GR–CNT/PP Improvements in Literature	Representative References
Elastic modulus (MPa)	~1367	~1518	+11%	Typical CNT/PP or GR–CNT/PP improvements in literature: ~10–40% increase in elastic modulus, typically at 0.1–3 wt% CNT loadings	Bao and Tjong 2008; Bikiaris et al. 2008; Choi et al. 2009; Prashantha et al. 2009
Crystallinity Xc (%)	~46	~46	≈ same	MWCNTs generally exhibit a modest nucleating effect in PP, leading to slight increases in crystallinity (~37 → 41–42%) or, in some systems, no significant change	Tabuani et al. 2007; Xu and Wang 2008; Bhuiyan et al. 2013
Melting Temperature (Tm) (°C)	163	162	≈ same	Tm values (~161–163 °C) are consistent with recent PP/MWCNT studies, where the addition of MWCNTs has a negligible influence on the melting peak temperature	Han et al. 2018; Stanciu et al., 2021; Bata et al. 2025
TGA onset (°C)	~423	~424	≈ same	Low MWCNT loadings generally have negligible effect on the onset of thermal degradation of PP, with reported shifts within a few degrees (typical onset ~380–420 °C).	Ávila-Orta et al., 2013; Naddeo et al., 2017; Han et al., 2018;
End Temp (°C)	~469	~469	≈ same	Main degradation step ending around 460–480 °C is consistent with reported Tmax values for PP and PP/CNT-based nanocomposites	Han et al., 2018; Cabello-Alvarado et al., 2019
Mass Loss (%)	~99	~97	≈ same	Low-loading MWCNT/PP nanocomposites generally exhibit only minor decreases in mass loss (increase in residue) upon TGA, consistent with ~97-99% mass loss observed in this study.	Kashiwagi et al., 2002; Zhou et al., 2016
DTG Peak Temp (°C)	~451	~452	+1 °C (small)	At very low CNT loadings (≈0.01 wt%), DTG peak temperatures typically remain close to neat PP. Significant increases in DTG peak temperature are generally observed only at higher CNT contents (≥1–5 wt%), with reported shifts of +10 to +35 °C.	Kashiwagi et al., 2004; Sahli et al., 2020

was integrated into a thermoplastic polypropylene matrix for the first time using the coagulation method. The surface functionalization of that filler was accomplished through oxidation and silanization.

Raman spectroscopy identified structural defects and confirmed successful modifications. Among the composite materials, the 0.01 wt% silane-modified GR-doped MWCNT/PP film showed the best mechanical performance. Improved interfacial interaction and good dispersion increased the elastic modulus and tensile strength. At higher filler levels, agglomeration occurred, resulting in a reduction in the mechanical performance.

Thermal analyses (DSC, TGA, DTG) indicated that modified GR-doped MWCNTs did not significantly affect the melting temperature or thermal degradation mechanism of PP. The crystallinity increased by ≈3% at 0.05 wt% filler loading, while no meaningful shift was noted in T_{onset} or T_{max} compared with neat PP. SEM images confirmed even particle dispersion at low filler levels, which directly relates to the observed mechanical improvements.

With minimal filler content, the addition of silane-functionalized GR-doped MWCNTs led to ≈11% and 48% increase in elastic modulus and tensile strength parameters, respectively, without noticeable changes in melting or crystallization characteristics. This performance profile is suitable for lightweight structural applications that require high mechanical strength and stable thermal performance, such as interior automotive parts, small casings for electronic devices, and flexible packaging exposed to elevated temperatures.

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Author contribution: Seçil Çelik Erbaş: planned and designed the study, wrote the article by analyzing the study, Zehra Üçtepe and Betül Soytürk: Gathered and analyzed data about the study, S.Bahar Baştürk: wrote the article by analyzing the study.

Ethics committee approval: If available, ethics committee approval should be added. The institution giving the ethics committee approval, approval number, and date should be written on.

References

Altay, L., Sarıkanat, M., Sağlam, M., Uysalman, T., & Seki, Y. (2021). The effect of various mineral fillers on thermal, mechanical, and rheological properties of polypropylene. Research on Engineering Structures & Materials, 7(3), 361-373. https://doi.org/10.17515/resm2021.258ma0213

Atif, R., & Inam, F. (2016). Reasons and remedies for the agglomeration of multilayered graphene and carbon nanotubes in polymers. Beilstein Journal of Nanotechnology, 7, 1174–1196. https://doi.org/10.3762/bjnano.7.109

Ávila-Orta, C.A., Raudry-López, C.E., Dávila-Rodríguez, M.V., Aguirre-Figueroa, Y.A., Cruz-Delgado, V.J., Neira-Velázquez, M.G., …& Hsiao, B.S. (2013). Morphology, thermal stability, and electrical conductivity of polymer nanocomposites of isotactic polypropylene/multi-walled carbon nanotubes. International Journal of Polymeric Materials and Polymeric Biomaterials, 62(12), 635–641. https://doi.org/10.1080/00914037.2013.769159

Bao, S.P., & Tjong, S.C. (2008). Mechanical behaviors of polypropylene/carbon nanotube nanocomposites: The effects of loading rate and temperature. Materials Science and Engineering: A, 485(1–2), 508-516. https://doi.org/10.1016/j.msea.2007.08.050

Bata, A., Ronkay, F., Zhang, C., & Gerse, P. (2025). Effect of recycling on the thermal and rheological properties of PP/MWCNT composites used as liner materials. Polymers, 17(16), 2178. https://doi.org/10.3390/polym17162178

Bhuiyan, K.H., Rahman, M., Mina, F., Islam, M.R., Gafur, A., & Begum, A. (2013). Crystalline morphology and properties of multi-walled carbon nanotube filled isotactic polypropylene nanocomposites: influence of filler size and loading. Composites Part A: Applied Science and Manufacturing, 52, 70-79. https://doi.org/10.1016/j.compositesa.2013.05.011

Bikiaris, D. (2010). Microstructure and properties of polypropylene/carbon nanotube nanocomposites. Materials, 3(4), 2884-2946. https://doi.org/10.3390/ma3042884

Bose, S., & Bandyopadhyay, A. (eds.) (2017). Materials and devices for bone disorders. 1st. Edition, Academic Press, Amsterdam, 543 pp.

Caban, R. (2022). FTIR-ATR spectroscopic, thermal and microstructural studies on polypropylene-glass fiber composites. Journal of Molecular Structure, 1264(4), 133181. https://doi.org/10.1016/j.molstruc.2022.133181

Cabello-Alvarado, C., Reyes-Rodríguez, P., Andrade-Guel, M., Cadenas-Pliego, G., Pérez-Alvarez, M., Cruz-Delgado, V.J.,…& Ávila-Orta, C.A. (2019). Melt-mixed thermoplastic nanocomposite containing carbon nanotubes and titanium dioxide for flame retardancy applications. Polymers, 11(7), 1204. https://doi.org/10.3390/polym11071204

Campo, E.A. (2008). Selection of polymeric materials: How to select design properties from different standards, William Andrew Publishing, New York, 253 pp.

Chiu, W.M., & Chang, Y.A. (2008). Chemical modification of multiwalled carbon nanotube with the liquid phase method. Journal of Applied Polymer Science, 107, 1655–1660. https://doi.org/10.1002/app.26633

Choi, S., Jeong, Y., Lee, G.W., & Cho, D.H. (2009). Thermal and mechanical properties of polypropylene filaments reinforced with multiwalled carbon nanotubes via melt compounding. Fibers and Polymers, 10, 513–518. https://doi.org/10.1007/s12221-009-0513-y

Coppola, B., Maio, L.D., Incarnato, L., & Tulliani, J.M. (2020). Preparation and characterization of polypropylene/carbon nanotubes (PP/CNTs) nanocomposites as potential strain gauges for structural health monitoring. Nanomaterials, 10(4), 814. https://doi.org/10.3390/nano10040814

Datsyuk, V., Kalyva, M., Papagelis, K., Parthenios, J., Tasis, D., Siokou, A.,… & Galiotis, C. (2008). Chemical oxidation of multiwalled carbon nanotubes. Carbon, 46, 833-840. https://doi.org/10.1016/j.carbon.2008.02.012

Deng, H., Reynolds, C.T., Cabrera, N.O., Barkoula, N.M., Alcock, B., & Peijs, T. (2010). The water absorption behaviour of all-polypropylene composites and its effect on mechanical properties. Composites Part B: Engineering, 41(4), 268-275. https://doi.org/10.1016/j.compositesb.2010.02.007

Gao, C., Guo, M., Liu, Y., Zhang, D., Gao, F., Sun, L., …& Wang, Y. (2023). Surface modification methods and mechanisms in carbon nanotubes dispersion. Carbon, 212, 118133. https://doi.org/10.1016/j.carbon.2023.118133

Gopalakrishnan, K., Moses, K., Dubey, P., & Rao, C.N.R. (2012). A Raman study of the interaction of electron-donor and -acceptor molecules with chemically doped graphene. Journal of Molecular Structure, 1023, 2–6. https://doi.org/10.1016/j.molstruc.2011.12.031

Greene, JP. (2021). Automotive plastics and composites: Materials and processing. William Andrew Publishing, Oxford, 392 pp. https://doi.org/10.1016/C2018-0-03030-3

Guarino, V., & Ambrosio, L. (eds.) (2018). Electrofluidodynamic technologies (EFDTs) for biomaterials and medical devices: Principles and advances. Woodhead Publishing, Duxford, 426 pp.

Han C., Sahle-Demessie E., Zhao A.Q., Richardson T., & Wang J. (2018). Environmental aging and degradation of multiwalled carbon nanotube reinforced polypropylene. Carbon, 129, 137-151. https://doi.org/10.1016/j.carbon.2017.10.038

Hussein, S.I., Ali, N.A., Khalil, A.S., Muslam, Z.R., & Jawad, M.K. (2024). Enhancement electrical and thermal conductivity of polypropylene doped MWCNT in energy materials application. AIP Conference Proceedings, 3229 (1), 070017. https://doi.org/10.1063/5.0236824

Imran, K.A., Lou, J., & Shivakumar, K.N. (2018). Enhancement of electrical and thermal conductivity of polypropylene by graphene nanoplatelets. Journal of Applied Polymer Science, 135(9), 45833. https://doi.org/10.1002/app.45833

Karian, H. (Ed.). (2003). Handbook of polypropylene and polypropylene composites, revised and expanded (Second edition). CRC Press, New York, 576 pp. https://doi.org/10.1201/9780203911808

Kashiwagi,T., Grulke, E., Hilding, J., Harris, R., Awad, W., & Douglas, J. (2002). Thermal degradation and flammability properties of poly(propylene)/carbon nanotube composites. Macromolacular Rapid Communications, 23, 761–765. https://doi.org/10.1002/1521-3927(20020901)23:13%3C761::AID-MARC761%3E3.0.CO;2-K

Kashiwagi, T., Grulke, E., Hilding, J., Groth, K., Harris, R., Butler, K.,…& Douglas, J. (2004). Thermal and flammability properties of polypropylene/carbon nanotube nanocomposites. Polymer, 45(12), 4227-4239. https://doi.org/10.1016/j.polymer.2004.03.088

Kathi, J., & Rhee, K.Y. (2008). Surface modification of multiwalled carbon nanotubes using 3-aminopropyltriethoxysilane. Journal of Materials Science, 43, 33–37. https://doi.org/10.1007/s10853-007-2209-2

Lavorgnaa, M., Romeoa V., Martonea, A., Zarrellia, M., Giordanoa, M., Buonocorea, G.G. …,& Xiac, H.S. (2013). Silanization and silica enrichment of multiwalled carbon nanotubes: Synergistic effects on the thermal-mechanical properties of epoxy nanocomposites. European Polymer Journal, 49(2), 428-438. https://doi.org/10.1016/j.eurpolymj.2012.10.003

Lee, J.H., Kathi, J., Rhee, K.Y., & Lee, J.H. (2010). Wear properties of 3-Aminopropyltriethoxysilane-functionalized carbon nanotubes reinforced ultra high molecular weight polyethylene nanocomposites. Polymer Engineering and Science, 50(7), 1433-1439. https://doi.org/10.1002/pen.21682

Leelapornpisit, W., Ton-That, M.T., Perrin-Sarazin, F., Cole, K.C., Denault, J., & Simard, B. (2005). Effect of carbon nanotubes on the crystallization and properties of polypropylene. Journal of Polymer Science Part B: Polymer Physics, 43(18), 2445 – 2453. https://doi.org/10.1002/polb.20527

Liu, B., Sun, J., Zhao, J., & Yun, X. (2025). Hybrid graphene and carbon nanotube–reinforced composites: Polymer, metal, and ceramic matrices. Advance Composites and Hybrid Materials, 8(1). https://doi.org/10.1007/s42114-024-01074-3

Luo, G., Liu, G., Chen, Y., Liang, W., Liu, G., Niu, Y., & Li, G. (2018). High performance glass fiber reinforced polypropylene realized by reactive extrusion technology. Composites Science and Technology, 165, 198-205. https://doi.org/10.1016/j.compscitech.2018.07.009

Ma, P.C., Siddiqui N.A., Marom, G., & Kim J.K. (2010). Dispersion and functionalization of carbon nanotubes for polymer-based nanocomposites: A review. Composites Part A: Applied Science and Manufacturing, 41(10), 1345-1367. https://doi.org/10.1016/j.compositesa.2010.07.003

Mark, J.E. (ed.). (1996). Physical properties of polymers handbook. Springer, New York, 1076 pp.

Mashhadzadeh, A.H., Fereidoon, Ab., & Ahangari, M.G. (2017). Surface modification of carbon nanotubes using 3-aminopropyltriethoxysilane to improve mechanical properties of nanocomposite based polymer matrix: experimental and density functional theory study. Applied Surface Science, 420, 167-179. https://doi.org/10.1016/j.apsusc.2017.05.148

Mazov, I.N., Kuznetsov, V.L., Krasnikov, D.V., Rudina, N.A., Romanenko, A.I., Anikeeva, O.B. …, & Zhuravlev V.A. (2011). Structure and properties of multiwall carbon nanotubes/polystyrene composites prepared via coagulation precipitation technique. Journal of Nanotechnology, 2011, 1-7. https://doi.org/10.1155/2011/648324

Merter, N.E. (2009). Effects of processing parameters on the mechanical behavior of continuous glass fiber/polypropylene composites. Master of Science Thesis, İzmir Institute of Technology, 92 pp.

Murray, R., Burke, M., Iacopino, D., & Quinn, A. J. (2021). Design of experiments and optimization of laser-induced graphene. ACS Omega, 6 (26), 16736-16743. https://doi.org/10.1021/Acsomega.1c00309

Muzyka, R., Drewniak, S., Pustelny, T., Chrubasik, M., & Gryglewicz, G. (2018). Characterization of graphite oxide and reduced graphene oxide obtained from different graphite precursors and oxidized by different methods using Raman spectroscopy. Materials, 11(7), 1050. https://doi.org/10.3390/ma11071050

Naddeo, C., Vertuccio, L., Barra, G., & Guadagno, L. (2017). Nano-charged polypropylene application: realistic perspectives for enhancing durability. Materials, 10(8), 943. https://doi.org/10.3390/ma10080943

Novikov, I.V., Krasnikov, D.V., Vorobei, A.M., Zuev, Y.I., Butt, H.A., Fedorov, F.S., ...& Nasibulin, A.G. (2022). Multifunctional elastic nanocomposites with extremely low concentrations of single-walled carbon nanotubes. ACS Applied Materials&Interfaces, 14(16), 18866-18876. https://doi.org/10.1021/acsami.2c01086

Omar, M.S.M., Lin, O.H., Akil, H.Md., Azza, N.A.N.N., Safwan, M.M., & Sofiah M.K.A. (2014). Thermal properties of polypropylene/palm kernel shell biocomposites: effects of amino silane (APTES). Materials Science Forum, 803, 250-254. https://doi.org/10.4028/www.scientific.net/MSF.803.250

Pang, A.L., & Ismail, H. (2013). Effects of kenaf loading and 3-aminopropyltriethoxysilane coupling agent on the properties of polypropylene/waste tire dust/kenaf composites. Journal of Thermoplastic Composite Materials, 27, 1607-1619. https://doi.org/10.1177/0892705712475002

Piao, Y., Tondare, V.N., Davis, C.S., Gorham, J.M., Petersen, E.J., Gilman, J.W., …& Walker, A.R.H. (2021). Comparative study of multiwall carbon nanotube nanocomposites by Raman, SEM, and XPS measurement techniques. Composites Science and Technology, 208, 108753. https://doi.org/10.1016/j.compscitech.2021.108753

Prashanthaa, K., Soulestina, J., Lacrampea, M.F., Krawczaka, P., Dupinb, G., & Claes M. (2009). Masterbatch-based multi-walled carbon nanotube filled polypropylene nanocomposites: Assessment of rheological and mechanical properties. Composites Science and Technology, 69, 1756-1763. https://doi.org/10.1016/j.compscitech.2008.10.005

Sahebian, S., Zebarjad, S.M., vahdati Khaki, J., & Lazerri, A. (2015). A study on the dependence of structure of multiwalled carbon nanotubes on acid treatment. Journal of Nanostructure in Chemistry, 5, 287–293. https://doi.org/10.1007/s40097-015-0160-3

Sahli, M., Barrière, T., Roazard, X., & Assoul, M. (2020). Investigating mechanical, thermal and rheological properties of polypropylene/carbon nanotubes composites. Microsystem Technologies, 26, 3023–3027. https://doi.org/10.1007/s00542-020-04833-6

Salihu, A.G., Thomas, S.P., Atieh, M.A., Mezghani, K., De, S.K., Bandyopadhyay, S., & Al-Juhani A. (2011). Effect of –COOH functionalized carbon nanotubes on mechanical, dynamic mechanical and thermal properties of polypropylene nanocomposites. Journal of Thermoplastic Composite Materials, 25(3), 333-350. https://doi.org/10.1177/0892705711406159

Shirvanimoghaddam, K., Balaji, K.V., Yadav, R., Zabihi, O., Ahmadi, M., Adetunji, P., & Naebe, M. (2021). Balancing the toughness and strength in polypropylene composites. Composites Part B: Engineering, 223, 109121. https://doi.org/10.1016/j.compositesb.2021.109121

Sin, L.T., & Tueen, B.S. (2023). Plastics and sustainability: Practical approaches. Elsevier, Amsterdam, 318 pp.

Stanciu, N.V., Stan, F., Sandu, I.L., Fetecau, C., & Turcanu, A.M. (2021). Thermal, rheological, mechanical, and electrical properties of polypropylene/multi-walled carbon nanotube nanocomposites. Polymers, 13(2), 187. https://doi.org/10.3390/polym13020187

Tabuani, D., Granelli, W., Camino, G., & Claes, M. (2007). Polypropylene based carbon nanotubes composites: Structure and properties. e-Polymers, 8(1), 1–12. https://doi.org/10.1515/epoly.2008.8.1.1178

Tsioptsias, C., Leontiadis, K., Ntampou, X., & Tsivintzelis, I. (2024). Modification of talc and mechanical properties of polypropylene-modified talc composite drawn fibers. Journal of Composites Science, 8(3), 91. https://doi.org/10.3390/jcs8030091

Vast, L., Philippin, G., Destrée, A., Moreau, N., Fonseca, A., Nagy, J.B., …& Mekhalif, Z. (2004). Chemical functionalization by a fluorinated trichlorosilane of multi-walled carbon nanotubes. Nanotechnology, 15(7), 781. https://doi.org/10.1088/0957-4484/15/7/011

Wang, J., Song, F., Ding, Y., & Shao, M. (2020). The incorporation of graphene to enhance mechanical properties of polypropylene self-reinforced polymer composites. Materials & Design, 195, 109073. https://doi.org/10.1016/j.matdes.2020.109073

Xu, D., & Wang, Z. (2008). Role of multi-wall carbon nanotube network in composites to crystallization of isotactic polypropylene matrix. Polymer, 49(1), 330–338. https://doi.org/10.1016/j.polymer.2007.11.041

Yang, B.X., Shi, J.H., Pramoda, K.P., & Goh, S.H. (2008). Enhancement of the mechanical properties of polypropylene using polypropylene-grafted multiwalled carbon nanotubes. Composites Science and Technology, 68(12), 2490-2497. https://doi.org/10.1016/j.compscitech.2008.05.001

Yu, W., Sisi, L., Haiyan, Y., & Jie, L. (2020). Progress in the functional modification of graphene/graphene oxide: A Review. RSC Advances, 10(26), 15328-15345. https://doi.org/10.1039/d0ra01068e

Zeinedini, A., Akhavan-Safar, A., & da Silva L.F.M. (2026). The role of agglomeration in the physical properties of CNTs/polymer nanocomposites. The Journal of Materials, 240(2), 193-231. https://doi.org/10.1177/14644207251316470

Zhou, T.Y., Tsui, G.C.P., Liang, J.Z., Zou, S.Y., Tang, C.Y., & Mišković-Stanković, V. (2016). Thermal properties and thermal stability of PP/MWCNT composites. Composites Part B: Engineering, 90, 107-114. https://doi.org/10.1016/j.compositesb.2015.12.013