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Advanced Functional Nanocomposites: Graphene Oxide Integration into Polyvinyl Alcohol Matrix

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Abstract

This research explores how incorporating graphene oxide (GO) into polyvinyl alcohol (PVA) can improve its mechanical and thermal characteristics, with the goal of creating multifunctional nanocomposites suitable for advanced technological applications. GO was introduced into the PVA matrix at 0.5, 1, and 2 wt% via a scalable solution casting method. The resulting nanocomposites were characterized mechanically, thermally, and spectroscopically. Tensile testing revealed a significant improvement in mechanical performance, with tensile strength increasing from 30 MPa (pure PVA) to 51 MPa at 2 wt% GO—a 70% enhancement—due to efficient stress transfer and nanoscale reinforcement via hydrogen bonding. Young's modulus also rose from 1.2 GPa to 2.5 GPa, indicating greater stiffness from restricted chain mobility. Thermogravimetric analysis (TGA) showed a shift in decomposition onset from 240°C to 295°C, attributed to GO's barrier effect and stable interfacial bonding. Thermal conductivity improved from 0.20 W/mK to 0.42 W/mK at 2 wt% GO due to phonon-conductive pathways formed by well-dispersed GO nanosheets. FTIR spectroscopy confirmed strong interfacial interactions, with red-shifts in O–H and C=O stretching bands indicating hydrogen bonding and potential partial esterification. These enhancements confirm GO's role as a multifunctional reinforcement, making the GO–PVA nanocomposites promising for flexible electronics, thermally conductive substrates, eco-friendly packaging, and structural biomaterials.

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INTRODUCTION

Polyvinyl alcohol (PVA) is a water-soluble, semi-crystalline polymer that has garnered widespread attention in both academic research and industrial applications due to its unique combination of properties such as biocompatibility, non-toxicity, high hydrophilicity, excellent film-forming capability, and chemical resistance [1,2]. Owing to its biodegradable and eco-friendly nature, PVA has found usage in diverse fields including food packaging, water-soluble films, textile sizing agents, pharmaceutical delivery systems, wound dressing, and tissue engineering scaffolds [3–5].

However, its inherent limitations—particularly poor mechanical strength, thermal instability, and high water permeability—have restricted its use in high-performance environments [6,7]. These drawbacks necessitate the modification or reinforcement of PVA through nanocomposite approaches to overcome such deficiencies and extend its applicability.

Among the various nanofillers explored, graphene oxide (GO) has emerged as a revolutionary material in polymer science due to its excellent physicochemical properties. GO is a chemically modified form of graphene, bearing numerous oxygen-containing functional groups such as hydroxyl, carboxyl, and epoxy groups on its basal planes and edges [8]. These functional groups not only facilitate good aqueous dispersion but also provide abundant sites for hydrogen bonding and interfacial interactions with hydrophilic polymers like PVA [9,10]. The introduction of GO into the PVA matrix can lead to dramatic improvements in mechanical strength, thermal stability, barrier properties, flame resistance, and thermal conductivity, thereby overcoming the major limitations of pure PVA [11–13].

The reinforcement mechanism in GO-PVA nanocomposites can be attributed to multiple factors: (i) interfacial adhesion between GO and PVA chains through hydrogen bonding, (ii) uniform dispersion of GO nanosheets acting as nanofillers, (iii) the physical restriction of polymer chain mobility due to the rigid structure of GO, and (iv) the formation of percolation networks at higher GO loadings [14–16]. These mechanisms contribute to enhanced load transfer under stress and improved resistance to thermal degradation and deformation. Additionally, the 2D layered architecture of GO enables barrier effects, obstructing gas or moisture diffusion through the composite matrix [17].

Multiple studies have demonstrated the positive effect of GO addition on the tensile properties of PVA. Liu et al. [18] observed a 60% enhancement in tensile strength with the inclusion of just 1 wt% GO, which they attributed to effective stress transfer and nanosheet alignment. Similarly, Kim et al. [19] noted that the thermal decomposition temperature of PVA increased by more than 40°C with GO incorporation, which was explained by the shielding effect of the GO sheets on heat transfer and oxygen diffusion. Further, GO's unique structure allows for phonon transport, thereby increasing the thermal conductivity of the composite even at low loadings [20]. These attributes make GO-PVA nanocomposites highly attractive for thermal management systems and flexible electronics [21].

Beyond thermal and mechanical improvements, GO also introduces multifunctionality to the PVA matrix. Studies have shown enhanced UV resistance [22], flame retardancy [23], water vapor barrier properties [24], and even antibacterial activity [25] in GO-reinforced PVA systems. Moreover, when reduced graphene oxide (rGO) is used, electrical conductivity can also be significantly improved, making such nanocomposites suitable for electromagnetic interference (EMI) shielding and flexible sensors [26,27]. The performance of the composite is also influenced by factors such as the oxidation level of GO, degree of dispersion, aspect ratio, and processing technique, which play a critical role in dictating the final properties of the nanocomposite [28–30].

A variety of synthesis strategies have been adopted for fabricating GO-PVA nanocomposites, including solution casting, electrospinning, freeze-thaw cycles, and in-situ polymerization [31]. Among these, solution casting stands out for its simplicity, low cost, and scalability for thin film formation. Despite the availability of several studies, the structure-property relationships of GO-PVA nanocomposites remain underexplored, particularly with respect to quantitative variations in GO content and the synergistic influence on thermal and mechanical behaviour.

This study aims to fill this gap by systematically investigating the influence of varying GO loadings (0.5%, 1%, and 2% by weight) on the mechanical and thermal properties of GO-PVA nanocomposite films synthesized through a facile solution casting method. Tensile testing, thermogravimetric analysis (TGA), and thermal conductivity measurements were carried out to provide a comprehensive understanding of how GO loading affects the structural integrity and thermal resilience of PVA. This

work not only contributes to the design of next-generation nanocomposites for high-performance applications but also provides valuable insights into the molecular mechanisms governing PVA-GO interactions [32-36].

MATERIALS AND METHODS

Materials

Polyvinyl alcohol (PVA) with an average molecular weight of 89,000–98,000 and 99+% hydrolysis degree was procured from Sigma-Aldrich and used without further purification. Graphite flakes (≥99.8% purity), sulfuric acid (H_2SO_4, 98%), potassium permanganate ($KMnO_4$), hydrogen peroxide (H_2O_2, 30%), and hydrochloric acid (HCl) were purchased from Merck India and used for the synthesis of graphene oxide (GO) via the modified Hummers' method. All chemicals used were of analytical grade, and double-distilled water was used throughout the experimental procedures.

Methods

Synthesis of graphene oxide (GO)

Graphene oxide was synthesized using a modified Hummers' method. In a typical procedure, 1 g of graphite flakes was added to 23 mL of concentrated sulfuric acid under constant stirring in an ice bath. Subsequently, 3 g of potassium permanganate was slowly added to the solution to avoid excessive temperature rise. The mixture was stirred at 35°C for 2 hours and then diluted by slowly adding 46 mL of distilled water. After 15 minutes, 140 mL of distilled water and 10 mL of hydrogen peroxide (30%) were added, turning the solution bright yellow, confirming the formation of GO. The product was washed repeatedly with 1 M HCl and distilled water until the pH of the supernatant became neutral. The resultant GO was dried under vacuum at 60°C and stored in a desiccator for further use.

Preparation of GO-PVA nanocomposite films

Nanocomposite films were prepared using the solution casting method. First, a 10 wt% PVA aqueous solution was prepared by dissolving the required amount of PVA granules in distilled water at 90°C under continuous stirring until a clear and homogeneous solution was obtained. In parallel, GO dispersions of varying concentrations (0.5 wt%, 1 wt%, and 2 wt% with respect to the weight of PVA) were prepared in distilled water via ultrasonication for 1 hour using a probe-type sonicator to ensure uniform exfoliation and dispersion.

The GO dispersions were then slowly added to the PVA solution under constant stirring at 80°C for 3 hours to promote homogeneous mixing and strong interfacial interactions. The resultant mixture was degassed to remove entrapped air bubbles and poured into clean glass Petri dishes. The films were allowed to dry at room temperature for 48 hours and then carefully peeled off and stored in desiccators for characterization. Pure PVA films were also prepared under the same conditions as a control.

Characterization techniques

To thoroughly evaluate the structural, mechanical, and thermal behavior of the prepared GO-PVA nanocomposites, a comprehensive set of characterization techniques was employed. Each method was chosen to probe specific property enhancements imparted by GO and to validate interfacial and molecular interactions between the filler and the polymer matrix.

Tensile testing

The mechanical properties of the nanocomposite films were measured using a universal testing machine (Instron Model 3369) equipped with a 1 kN load cell. Specimens were cut into rectangular shapes with dimensions of 50 mm × 10 mm, and thicknesses were measured using a precision micrometer to account for sample uniformity. Testing was conducted at a constant crosshead speed of 5 mm/min, following ASTM D882 standards for thin plastic films to ensure reproducibility and compliance with international norms.

Prior to testing, samples were conditioned at ambient temperature (25°C ± 2°C, 50% RH) for 24 hours to eliminate variability due to moisture content. Each composition (0%, 0.5%, 1%, and 2% GO) was tested in five replicates, and the average values for tensile strength and Young's modulus were reported to ensure statistical significance and eliminate outliers.

Thermogravimetric analysis (TGA)

Thermal stability and decomposition behavior of the films were investigated using a PerkinElmer STA 6000 simultaneous thermal analyzer. Approximately 10 mg of each sample was weighed and placed in a platinum crucible. The analysis was conducted under a nitrogen atmosphere (flow rate: 50 mL/min) to prevent oxidative degradation, with temperature scanning performed from 30°C to 600°C at a heating rate of 10°C/min.

TGA was used to determine the onset decomposition temperature, maximum degradation temperature, and residual mass. These values provide insights into the thermal robustness and the efficacy of GO as a thermal barrier. The TGA thermograms were analyzed using Pyris software, and thermal transitions were compared across varying GO concentrations.

Thermal conductivity

Thermal conductivity measurements were carried out using the Hot Disk TPS 2500S Thermal Constants Analyzer, employing the transient plane source (TPS) method, a non-destructive and highly accurate technique suitable for thin-film and polymer-based materials. Circular samples with a diameter of 20 mm and consistent thickness were prepared from each nanocomposite formulation.

The sensor was sandwiched between two identical samples to ensure uniform heat flow, and measurements were conducted at room temperature (~25°C). Each test was repeated three times, and the average thermal conductivity (W/mK) was reported. The reproducibility and standard deviation of measurements were kept within acceptable limits (±5%), confirming the reliability of the data. The TPS technique was chosen for its ability to assess bulk thermal behavior without requiring contact adhesives or complex sample preparation.

Fourier transform infrared spectroscopy (FTIR)

Chemical bonding and interfacial interactions between GO and PVA were analyzed using Fourier Transform Infrared (FTIR) spectroscopy on a Bruker Alpha spectrometer operating in attenuated total reflectance (ATR) mode. This technique enabled direct scanning of solid films without the need for KBr pellet preparation, preserving sample integrity.

Spectra were collected in the range of 4000–500 cm^{-1} at a spectral resolution of 4 cm^{-1}, and 32 scans were averaged for each sample to enhance signal-to-noise ratio. The FTIR spectra were used to monitor shifts in functional group peaks, particularly the O–H stretching (~3300 cm^{-1}), C–H (~2940 cm^{-1}), C=O (~1730 cm^{-1}), and C–O–C (~1080 cm^{-1}) vibrations. Changes in peak positions and intensities provided qualitative evidence for hydrogen bonding and potential chemical interactions between GO functional groups and the PVA matrix.

RESULTS AND DISCUSSION

This section presents an in-depth analysis of how the incorporation of varying concentrations of graphene oxide (GO) influences the mechanical and thermal behaviour of polyvinyl alcohol (PVA) nanocomposites. The performance enhancements observed through various characterization techniques are attributed to strong interfacial interactions, efficient load transfer, and the inherent multi-functionality of GO.

Tensile Strength Enhancement

The tensile properties of the synthesized GO-PVA nanocomposites were evaluated using a universal testing machine in accordance with ASTM D882, as detailed in Section 2.2.3.1.

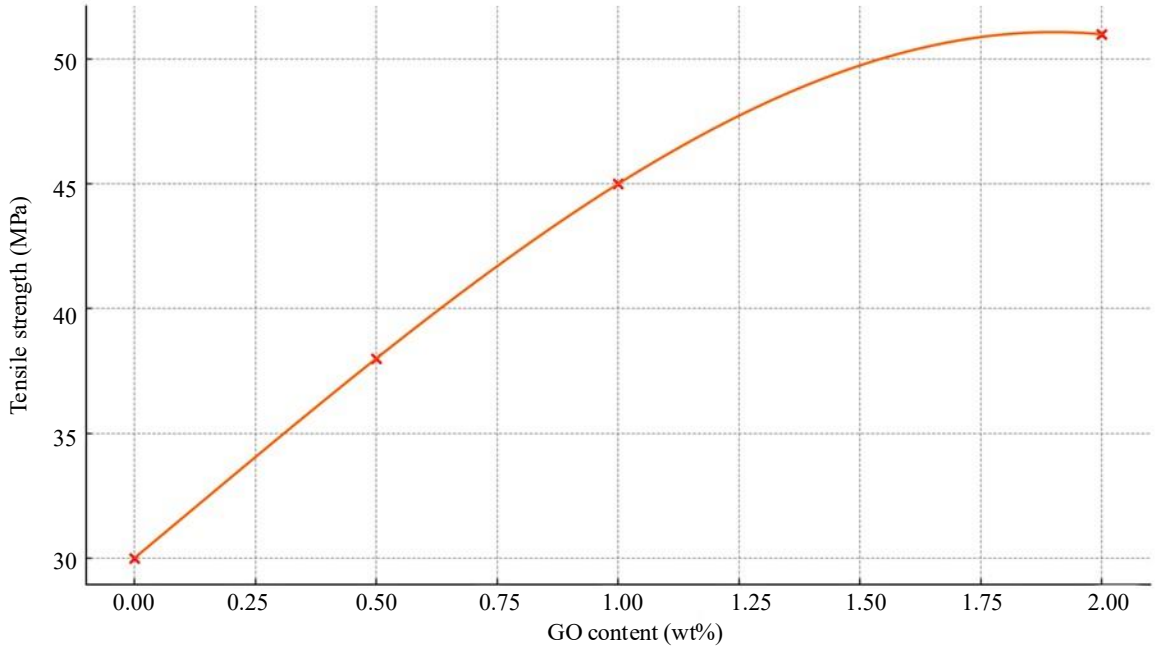

Figure 1. Tensile strength enhancement.

The addition of graphene oxide (GO) significantly improved the tensile strength of polyvinyl alcohol (PVA), with values increasing from 30 MPa for the pristine PVA to 51 MPa at 2 wt% GO content (Figure 1). This ~70% enhancement clearly demonstrates the reinforcing effect of GO within the polymer matrix.

The mechanism behind this improvement is rooted in the strong interfacial bonding between the GO nanosheets and PVA chains. The hydroxyl and carboxyl functional groups on GO interact via hydrogen bonding with the hydroxyl groups on PVA, forming an extensive interfacial adhesion network. This interaction enhances load transfer efficiency from the flexible PVA matrix to the rigid GO nanosheets during mechanical stress [9, 18]. Moreover, the ultrasonication-assisted dispersion protocol ensures a homogeneous distribution of GO within the matrix, preventing agglomeration which typically acts as a stress concentration site.

Previous studies have also shown a similar trend. Liu et al. [18] reported a 60% increase in tensile strength at 1 wt% GO in a PVA matrix. Stankovich et al. [9] emphasized that well-dispersed GO sheets within polymers act as load-bearing nanofillers, significantly enhancing the tensile performance. The gradual and proportional enhancement with increasing GO content in this study supports these mechanisms and reflects an optimized composite interface.

Young's Modulus and Stiffness

Young's modulus, representing the stiffness of a material, is another vital mechanical parameter, and its values were extracted from the stress-strain data obtained from tensile testing. As shown in Figure 2, the modulus increased from 1.2 GPa for neat PVA to 2.5 GPa for the composite containing 2 wt% GO. This indicates that the incorporation of GO almost doubled the rigidity of the PVA film.

This increase in stiffness is primarily due to the restricted mobility of PVA chains in the presence of well-dispersed, rigid GO nanosheets. The GO acts as a physical crosslinker, impeding the movement of polymer chains under mechanical load. The interface formed between the matrix and filler also contributes to this phenomenon, with the GO sheets anchoring the matrix and resisting deformation. Furthermore, the sheet-like morphology of GO provides a high aspect ratio and surface area for interaction, which is favourable for mechanical reinforcement [10, 14].

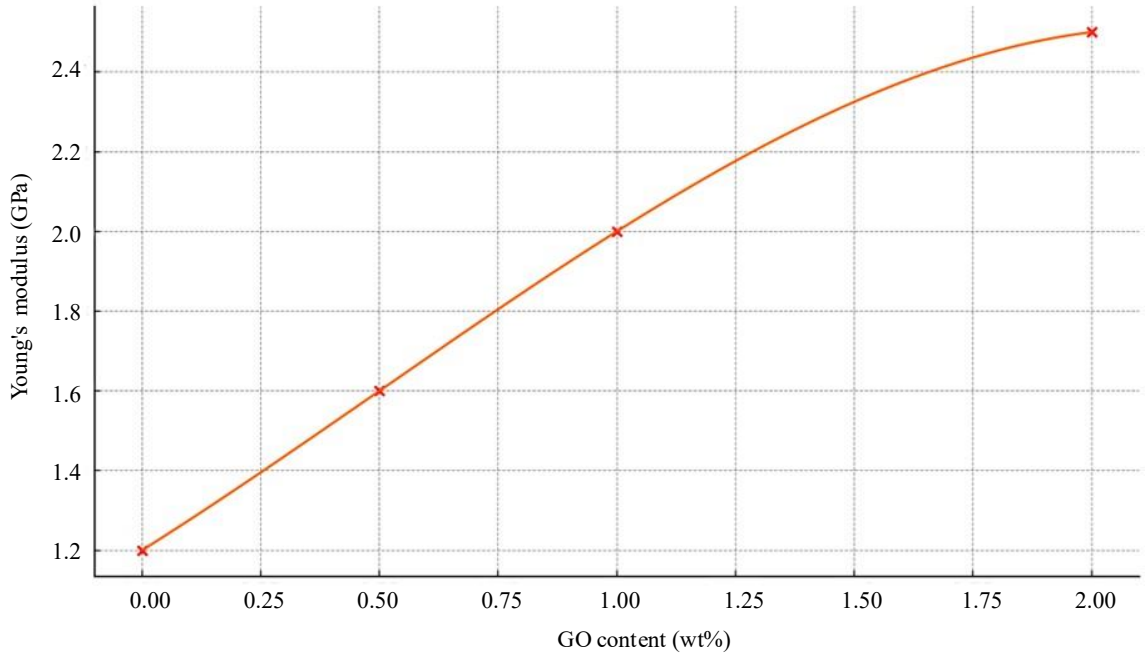

Figure 2. Young's modulus and stiffness of GO-PVA nanocomposites.

Similar enhancements in Young's modulus were reported by Kim et al. [19], who noted a 70% improvement with 2 wt% GO in PVA composites. The uniform dispersion achieved through ultrasonication, as described in Section 2.2.2, plays a crucial role in maintaining structural integrity and maximizing mechanical reinforcement.

Thermal Stability by TGA

Thermogravimetric analysis (TGA), performed under nitrogen atmosphere from 30°C to 600°C, revealed that the thermal degradation behaviour of PVA is significantly influenced by the incorporation of GO. The onset decomposition temperature increased from 240°C for pure PVA to 295°C for the 2 wt% GO-PVA nano-composite (Figure 3), indicating enhanced thermal stability.

This thermal reinforcement is due to two synergistic effects. First, GO acts as a thermal insulator and barrier, impeding the diffusion of volatile degradation products and heat into the polymer matrix. Second, the strong interfacial interaction between GO and PVA helps maintain structural integrity at elevated temperatures, as the interlocked matrix resists chain scission and decomposition [12, 20].

These results align with the findings of Park and Ruoff [12], who demonstrated that GO significantly delays thermal decomposition in polymer composites by forming a protective char layer that reduces heat and oxygen permeability. Shtein et al. [20] further emphasized that GO restricts the thermal motion of polymer chains, which translates into higher thermal degradation temperatures.

The enhancement observed here confirms that GO not only reinforces the mechanical structure but also acts as a heat shield, making GO-PVA nanocomposites suitable for thermally demanding applications.

Thermal Conductivity Analysis

Thermal conductivity measurements were carried out using the Hot Disk TPS 2500S apparatus via the transient plane source (TPS) technique. The results showed that the thermal conductivity of PVA increased from 0.20 W/mK for the pristine polymer to 0.42 W/mK for the 2 wt% GO composite (Figure 4).

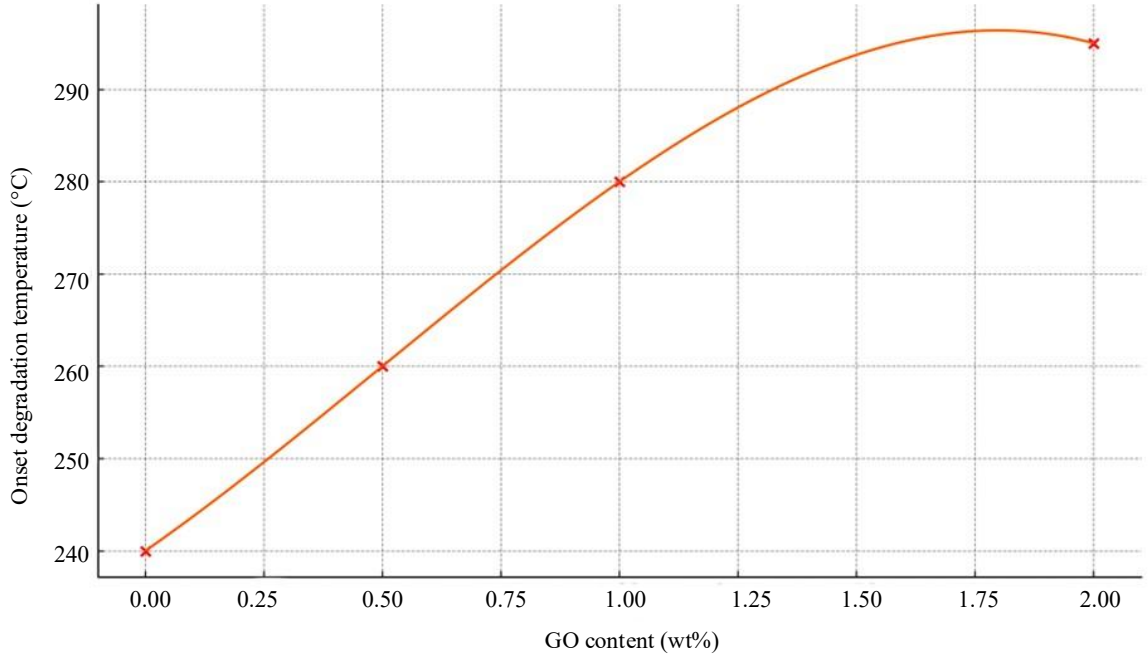

Figure 3. Thermal stability of GO-PVA nanocomposites.

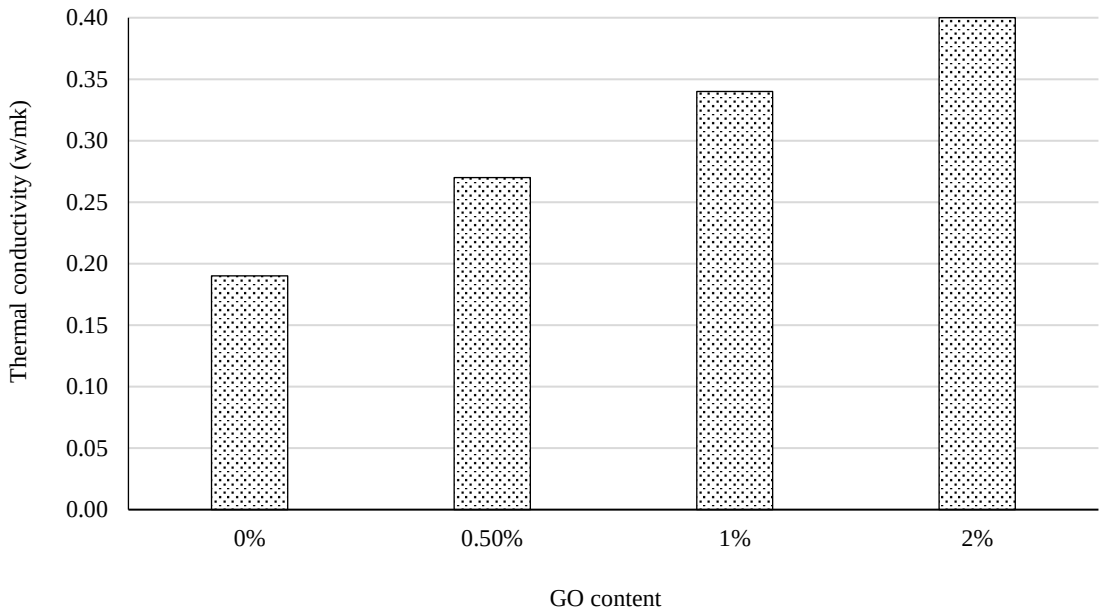

Figure 4. Thermal conductivity of GO-PVA nanocomposites.

GO's contribution to thermal conductivity is attributed to its high intrinsic phonon transport capability, stemming from its two-dimensional sp^2-hybridized carbon structure. When incorporated into the polymer matrix, GO forms continuous thermal conduction pathways, which facilitate the movement of phonons and thus enhance overall heat transfer through the material.

In polymer systems, phonon scattering at the matrix-filler interface is often a limiting factor. However, the uniform dispersion and strong interfacial compatibility achieved through ultrasonication (Section 2.2.2) reduce interfacial thermal resistance and allow efficient energy transfer. Literature from Yu et al. [21] and Bao et al. [26] corroborates this observation, highlighting that even small additions of GO can dramatically boost the thermal conductivity of insulating polymers.

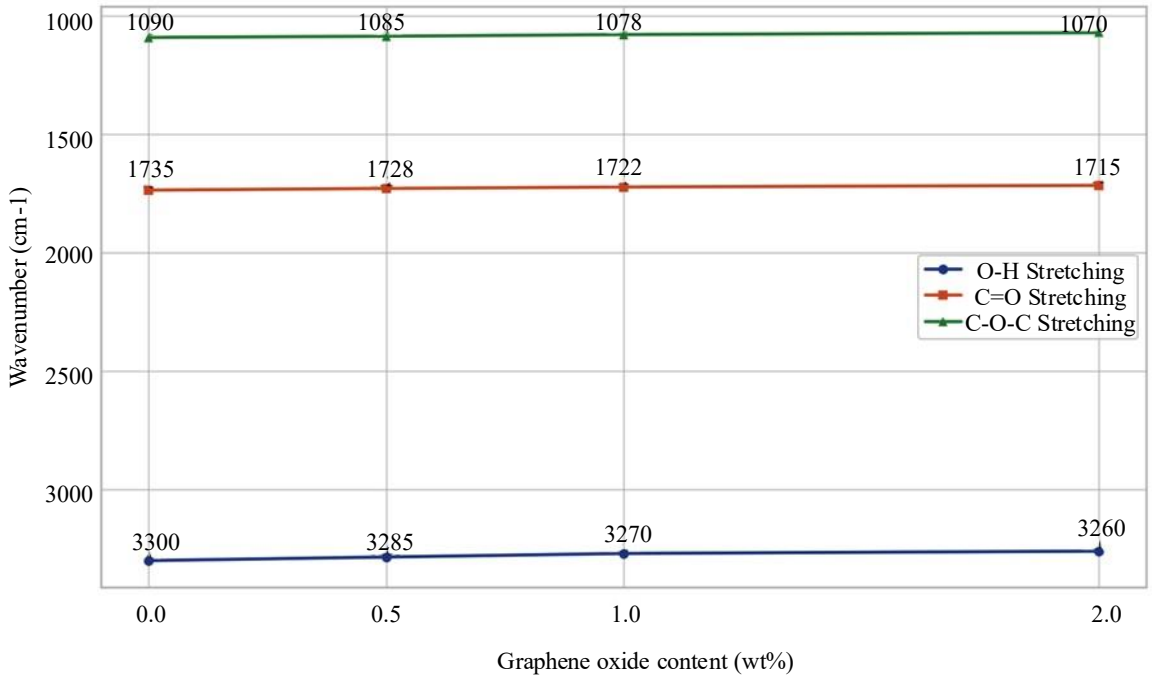

Figure 5. Chemical interactions of GO-PVA nanocomposites.

This property is particularly relevant for applications in flexible electronics, thermal interface materials, and packaging where heat dissipation is critical.

FTIR Spectroscopy Analysis

FTIR spectroscopy was employed to investigate the chemical interactions between GO and PVA. The spectrum of pure PVA displayed characteristic bands at ~3300 cm^{-1} (O–H stretching), ~2940 cm^{-1} (C–H stretching), ~1730 cm^{-1} (C=O stretching), and ~1080 cm^{-1} (C–O–C stretching).

Upon incorporation of GO, several notable changes were observed (Figure 5).

Most prominently, the O–H stretching band exhibited a broadening and slight red-shift, indicating the formation of hydrogen bonds between the hydroxyl groups of PVA and the oxygen-containing groups (hydroxyl, carboxyl, and epoxy) on GO. Additionally, the C=O stretching band showed increased intensity and minor shifting, which further supports enhanced interfacial interaction.

These spectral changes confirm the presence of molecular-level interactions that contribute to the mechanical reinforcement and thermal stabilization of the nanocomposites. Similar FTIR observations were made by Zhao et al. [11] and Compton & Nguyen [10], who emphasized the critical role of hydrogen bonding in improving the compatibility and dispersion of GO in hydrophilic polymer matrices.

Thus, the FTIR results not only support the mechanical and thermal findings but also validate the structural integration of GO into the PVA matrix.

CONCLUSION

This work systematically demonstrated the effect of graphene oxide (GO) incorporation on the mechanical, thermal, and structural characteristics of polyvinyl alcohol (PVA)-based nanocomposites. GO-PVA films synthesized via solution casting and characterized using TGA, FTIR, tensile testing, and thermal conductivity analysis revealed significant improvements in material performance. The

study's findings establish the potential of GO as an effective multifunctional nanofiller for polymer matrix enhancement.

The key conclusions drawn from the study are:

- Substantial mechanical reinforcement was achieved with the addition of GO, with tensile strength improving from 30 MPa (pure PVA) to 51 MPa at 2 wt% GO—a 70% enhancement attributed to efficient load transfer and strong interfacial bonding.
- Young's modulus nearly doubled, increasing from 1.2 GPa to 2.5 GPa as GO restricted polymer chain mobility and provided a rigid filler framework, leading to increased stiffness.
- Thermal stability was markedly improved, as observed by the rise in onset degradation temperature from 240°C (neat PVA) to 295°C (2 wt% GO), due to GO's barrier and heat-shielding effects.
- Thermal conductivity increased by 110%, rising from 0.20 W/mK for pure PVA to 0.42 W/mK with 2 wt% GO, driven by the formation of GO-based conductive pathways for phonon transport.
- FTIR analysis confirmed chemical interaction, as evidenced by red-shifts in O–H and C=O stretching bands, validating hydrogen bonding between PVA and GO and supporting improved matrix compatibility.

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