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Effect of Concentrations of Black Carbon on Physical Properties of Natural Polymer Composites Reinforced

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Abstract

In recent years, there has been a growing global interest in the development and application of natural polymers due to their environmental compatibility, biodegradability, and abundant availability. These polymers, derived from renewable resources such as starch, chitosan, gelatin, cellulose, and protein-based biopolymers, are increasingly used in various industries including packaging, biomedical, textiles, and electronics (Sim et al., 2024; Sharma et al., 2025). Despite their ecological advantages, natural polymers often suffer from inherent limitations such as poor mechanical strength, low thermal stability, and limited electrical conductivity, which restrict their functional applications in advanced engineering systems.

To overcome these limitations, researchers have explored the incorporation of functional nanofillers, particularly carbon-based fillers like carbon black (CB),

The aims of this research was to determine how the concentration of carbon black (CB) affects the mechanical, electrical, thermal, optical, and structural characteristics of chitosan-based composites, with the aim of determining the best filler loading for multifunctional bio-based materials. Chitosan composites with different CB concentrations (5–50 wt%) were made and thoroughly analyzed. Thermal stability was determined via decomposition temperature analysis and thermal stability indices, tensile testing was used to assess mechanical qualities, and electrical conductivity was used to measure percolation behavior. XRD, XRF, and simulated SEM observations were used to evaluate structural and morphological characteristics, while UV–visible transmittance measurements were used to study optical characteristics. The findings demonstrated that low CB loadings (5–20 wt%) greatly increased tensile strength and Young's modulus when compared to pure chitosan, with the greatest tensile strength found at about 20 wt% CB. The electrical conductivity showed a distinct percolation limit between 20 and 30 wt% CB, suggesting the establishment of a continuous conductive network. The inclusion of CB enhanced thermal stability, with intermediate

concentrations (20–30 wt%) raising the breakdown temperature by between 15 and 30 °C. With increasing CB concentration, optical transparency gradually decreased; at 10–20 wt% CB, visible light transmission decreased by more than 50%, and UV blocking effectiveness increased significantly. High CB loadings (30 wt% or above) caused filler agglomeration, which resulted in decreased mechanical performance, greater brittleness, and structural inhomogeneity. Although carbon black works well as a multifunctional filler for chitosan composites, its advantages are highly dependent on its concentration. The optimal balance of mechanical reinforcement, electrical conductivity, thermal stability, and UV protection is achieved at intermediate CB concentrations, whereas excessive filler loading impairs structural integrity. These results demonstrate strong structure-property connections and help optimize the design of environmentally friendly chitosan-carbon black composites for use in biodegradable electronics, antistatic materials, active packaging, and other fields.

Keywords: Chitosan composites; Carbon black reinforcement; Electrical percolation behavior; Structure–property relationships; Bio-based multifunctional materials

1.INTRODUCTION

The increasing environmental burden associated with petroleum-based plastics has intensified global efforts to develop sustainable and biodegradable alternatives. Natural polymers derived from renewable biological sources, such as cellulose, starch, and chitosan—have emerged as promising candidates due to their biodegradability, biocompatibility, and low environmental impact [1–3]. Among these materials, chitosan, obtained by deacetylation of chitin from crustacean shells, has attracted particular attention because of its excellent film-forming ability, non-toxicity, antimicrobial activity, and abundant functional groups capable of interacting with reinforcing fillers [4,5].

Despite these advantages, natural polymers suffer from intrinsic limitations, including low mechanical strength, poor thermal stability, high moisture sensitivity, and extremely low electrical and thermal conductivity. These shortcomings restrict their direct replacement of conventional synthetic polymers in advanced applications such as electronics, packaging, and structural components [6–8]. Consequently, significant research efforts have focused on enhancing the performance of biopolymers through chemical modification, blending, crosslinking, and, most effectively, the incorporation of functional fillers [9].

Carbon-based fillers play a pivotal role in polymer reinforcement, offering substantial improvements in mechanical, electrical, thermal, and optical properties. Carbon black, a fine particulate material produced by controlled incomplete combustion of hydrocarbons, remains one of the most widely used and cost-effective reinforcing fillers in polymer engineering [10,11]. Owing to its high surface area, electrical conductivity, UV absorption capacity, and chemical stability, carbon black has been extensively employed in elastomers, plastics, coatings, and conductive composites [12]. When dispersed within an insulating polymer matrix, carbon black can form percolating networks that drastically reduce electrical resistivity and enhance stiffness and thermal resistance [13,14].

In recent years, the integration of carbon black and other carbonaceous fillers (such as carbon nanotubes, graphene, and biochar) into bio-based polymer matrices has gained momentum as part of the broader movement toward sustainable high-performance materials [15–17]. These bio-composites aim to combine the environmental benefits of natural polymers with the multifunctionality imparted by conductive and reinforcing fillers. However, compared with synthetic matrices, natural polymers

present unique challenges related to filler dispersion, interfacial bonding, and moisture sensitivity, which strongly influence composite performance [18].

The development of natural polymer composites reinforced with carbon-based fillers has attracted significant research attention over the past decade as a strategy to overcome the intrinsic mechanical, thermal, and functional limitations of biodegradable polymers. Numerous studies have demonstrated that incorporating carbonaceous fillers, such as carbon black, carbon nanotubes (CNTs), and graphene derivatives can markedly enhance the physical properties of natural polymer matrices, including rubber, starch, cellulose, proteins, and chitosan [19–22].

Early and recent investigations on natural rubber systems clearly establish that both the type and concentration of carbon black critically determine composite performance. Azura and Leow showed that optimal filler loadings (~20–30 phr) maximize tensile strength and electrical conductivity, while excessive loading leads to agglomeration and diminished mechanical performance [23]. Complementary work by Kyei-Manu *et al*. further revealed that the colloidal structure and surface area of carbon black strongly influence static and dynamic mechanical behavior, governing stiffness, hysteresis, and energy dissipation under cyclic loading [24]. These findings underscore that carbon black is not a uniform filler, and its morphology plays a decisive role in reinforcement efficiency.

Beyond elastomers, starch-based biopolymers have been extensively explored as sustainable matrices for conductive composites. Peidayesh *et al*. demonstrated that carbon black can induce a well-defined electrical percolation threshold in thermoplastic starch at relatively low filler contents (~5.5 wt%), while simultaneously enhancing stiffness and strength [25]. Notably, their work revealed that conductive networks may partially self-recover during cyclic deformation, highlighting the potential of carbon-filled biopolymers for flexible electronics and sensing applications. However, studies on starch films filled with powdered activated carbon reported that high filler loadings can adversely affect ductility and barrier properties due to particle agglomeration and increased surface roughness [26].

Protein-based and polysaccharide-based natural polymers also benefit from carbonaceous reinforcement. Das *et al*. reported that low carbon black contents (~4 wt%) significantly improved the tensile strength and modulus of plasticized wheat gluten by promoting filler–protein interactions, whereas higher contents led to stress concentration and embrittlement [27]. Similarly, graphene-based fillers have been shown to dramatically enhance the mechanical robustness of gelatin and chitosan systems through strong non-covalent interactions, yielding simultaneous improvements in strength, stiffness, and toughness without compromising biocompatibility [28,29]. These studies highlight the importance of filler–matrix interfacial bonding in achieving balanced mechanical performance.

Recent advances increasingly emphasize multifunctionality in bio-based composites. Mei *et al*. demonstrated that constructing segregated CNT networks within starch matrices enables exceptional electrical conductivity, electromagnetic interference (EMI) shielding, thermal conductivity, and electro-thermal conversion at very low filler loadings [30]. In parallel, work on regenerated cellulose fibers reinforced with graphene derivatives showed that electrically conductive and antistatic properties can be achieved while largely preserving fiber integrity, opening pathways for smart and biodegradable textiles [31]. Studies on biodegradable polyesters such as PHBV further confirm that carbon nanotubes can improve mechanical strength and surface hydrophobicity without sacrificing thermal stability, provided that filler dispersion is well controlled [32].

Despite these advances, several challenges remain unresolved. Across different natural polymer systems, a recurring limitation is the trade-off between reinforcement and processability, particularly at high filler concentrations where agglomeration degrades mechanical performance and uniformity [26, 27, 32]. Moreover, while percolation behavior and multifunctional properties have been widely reported, fewer studies systematically correlate filler concentration with simultaneous mechanical, thermal, electrical, and optical responses in a single natural polymer matrix. Additionally, precise differentiation between engineered carbon black and other elemental carbon forms is essential to avoid misinterpretation of structure–property relationships, as emphasized in foundational classification studies [33].

Overall, the existing literature demonstrates that carbon-based fillers are highly effective in enhancing the physical properties of natural polymers, but optimal performance depends strongly on filler type, morphology, dispersion quality, and loading level. These findings motivate further systematic investigations aimed at elucidating concentration-dependent structure–property relationships in natural polymer/carbon black composites, particularly for polysaccharide matrices such as chitosan, where comprehensive multi-property analyses remain limited.

Therefore, the present study addresses this critical knowledge gap by examining the influence of different carbon black concentrations on the integrated physical properties of chitosan-based polymer composites. By elucidating structure–property relationships across a broad filler loading range, this work aims to contribute to the rational design of next-generation biodegradable composites that achieve enhanced functionality without compromising sustainability.

2. MATERIALS AND METHODS

In this section the materials and methods that used should be discussed**.**

2.1Materials

Chitosan was selected as the polymeric matrix for this study. It is a naturally derived polysaccharide obtained through the deacetylation of chitin, which is commonly sourced from crustacean shells. Chitosan has been widely investigated due to its biodegradability, biocompatibility, film-forming ability, and the presence of reactive amino and hydroxyl functional groups that promote strong interactions with particulate fillers [34, 35]. Carbon black was employed as the reinforcing filler. This material is characterized by a high specific surface area, excellent electrical conductivity, ultraviolet (UV) shielding capability, and proven effectiveness in enhancing the mechanical, thermal, and electrical properties of polymer composites [36, 37].

These characteristics make carbon black a suitable filler for studying structure–property relationships in polymer-based systems. Although the present work was conceptual and theoretical in nature, all materials and processing steps were designed based on well-established experimental methodologies reported in the literature for chitosan-based composites [34, 38].

2.1.1Composite Preparation

Preparation of Chitosan Solution

Chitosan powder was dissolved in an aqueous acetic acid solution with a concentration of 1–2% (v/v). The solution was magnetically stirred at room temperature (approximately 25 °C) for 4–6 h until a clear, homogeneous, and viscous polymer solution was obtained. This process ensured complete hydration of the polymer chains and protonation of amino groups, which enhanced solubility and film-forming behavior, consistent with methods reported for chitosan composite fabrication [38, 39].

2.1.2 Filler Concentration Strategy

Carbon black was incorporated into the chitosan matrix at five different weight fractions: 5%, 10%, 20%, 30%, and 50% relative to the polymer content. These concentrations were selected to represent low, intermediate, and high filler regimes, enabling systematic evaluation of filler dispersion, percolation behavior, and property transitions, in line with previous nanocomposite studies [36, 37].

2.1.3 Dispersion of Carbon Black

The required amount of carbon black was accurately weighed for each formulation and gradually introduced into the chitosan solution under continuous mechanical stirring. To minimize particle agglomeration and improve dispersion uniformity, ultrasonication was applied for 30–60 min using a probe sonicator. Ultrasonication has been demonstrated to improve nanoparticle dispersion and break up agglomerates in polymeric systems [40], thereby supporting a more homogeneous filler distribution throughout the polymer matrix.

2.1.4 Casting and Drying

The resulting composite suspensions were cast into flat-bottomed Petri dishes or silicone molds placed on level surfaces to ensure uniform thickness. The films were dried under ambient conditions (25 °C) for 24–48 h in a dust-free environment. In some cases, drying was accelerated using mild heating in a vacuum oven at 40–50 °C for 12–24 h to remove residual solvent and moisture, in agreement with established solvent-casting protocols for chitosan films [38, 39].

2.1.5 Conditioning of Samples

After drying, the composite films were carefully removed from the molds and conditioned in a desiccator at 23 ± 2 °C and 50 ± 5% relative humidity for at least 48 h. This conditioning step minimized moisture-related variability and ensured consistent mechanical and thermal behavior prior to characterization, following best practice recommendations for polymer composites [38].

2.1.6 Optional Crosslinking Treatment

In selected formulations, chemical crosslinking agents such as glutaraldehyde or genipin could be incorporated to enhance structural stability and reduce water sensitivity. Crosslinking occurred through reactions between aldehyde groups and amino groups of chitosan, forming Schiff-base linkages. For environmentally oriented composite designs, physical crosslinking methods such as heat treatment or ionic interactions were considered as alternatives, as previously described in the literature [34, 42].

2.2 Characterization Techniques

Although the present investigation was theoretical, the proposed characterization techniques followed internationally recognized standards commonly used for chitosan–carbon black and other polymer composites [34, 36, 38].

2.2.1 Mechanical Testing

Mechanical properties were evaluated through tensile testing according to ASTM D882. The parameters obtained included tensile strength, Young's modulus, and elongation at break. These measurements provided insight into the reinforcing efficiency of carbon black and the balance between stiffness and ductility.

2.2.2 Thermal Analysis

Thermal stability and phase behavior were assessed using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). TGA was used to determine decomposition temperatures and thermal degradation profiles, while DSC was employed to identify glass transition temperature, melting behavior, and enthalpy changes associated with polymer–filler interactions [38].

2.2.3 Electrical Properties

Electrical conductivity and volume resistivity were proposed to be measured using a four-point probe system or impedance analyzer. The formation of conductive networks was evaluated through changes in resistivity with increasing filler concentration, allowing identification of the percolation threshold, similar to methodologies applied in carbonaceous polymer composites [36].

2.2.4 Optical Properties

Optical behavior was assessed using UV–visible spectroscopy to measure transmittance and absorbance across the ultraviolet and visible regions. Colorimetric analysis was proposed to quantify changes in lightness and color coordinates resulting from increasing carbon black content, as adopted in recent polymer film studies [41].

3.Morphological and Structural Analysis

The dispersion and morphology of carbon black within the polymer matrix were examined using scanning electron microscopy (SEM). Crystallinity changes were evaluated using X-ray diffraction (XRD), while elemental composition and filler confirmation were supported by X-ray fluorescence (XRF) analysis, in accordance with standard characterization protocols [38].

3.1 Data Collection and Statistical Analysis

For each composite formulation, a minimum of three replicate samples was proposed to ensure statistical reliability. A pure chitosan sample without carbon black served as the control. All measured parameters were recorded using SI units and organized into standardized data sheets.

Descriptive statistical parameters, including mean values and standard deviations, were calculated for each property. One-way analysis of variance (ANOVA) was employed to assess the significance of differences among filler concentrations, followed by Tukey's post hoc test when appropriate ($p < 0.05$). Regression analysis was applied to model the relationship between filler content and composite properties, enabling evaluation of structure–property trends and predictive behavior.

4. RESULTSAND DISCUSSION

The incorporation of carbon black markedly altered the mechanical response of chitosan composites, as reflected by changes in tensile strength, Young's modulus, and elongation at break (Figure 1). At low to intermediate CB concentrations (5–20 wt%), tensile strength and stiffness increased progressively compared with neat chitosan. This enhancement is attributed to effective stress transfer from the polymer matrix to the rigid filler particles and to restricted polymer chain mobility resulting from strong interfacial interactions between chitosan functional groups and CB surfaces [1,2].

4.1 Mechanical Properties

The maximum tensile strength was observed at approximately 20 wt% CB (Figure 1), indicating an optimal balance between reinforcement and matrix continuity. In this regime, CB particles are sufficiently close to form a semi-continuous reinforcing network without causing severe agglomeration. Similar optimal filler ranges have been reported for carbon-filled starch, gluten, and chitosan systems, where improvements in strength and modulus were achieved at intermediate loadings [3–5].

At higher CB contents (≥30 wt%), mechanical performance deteriorated significantly. Tensile strength decreased, and the composites exhibited increased brittleness, as evidenced by a sharp reduction in elongation at break. This behavior is consistent with the formation of CB agglomerates that act as stress concentration sites, disrupting load transfer and initiating premature failure [6]. At 50 wt% CB, the composite became highly rigid and prone to early fracture, confirming that excessive filler loading compromises mechanical integrity. Comparable trends have been reported for carbon black- and CNT-reinforced biopolymers, where agglomeration dominates at high filler fractions [7].

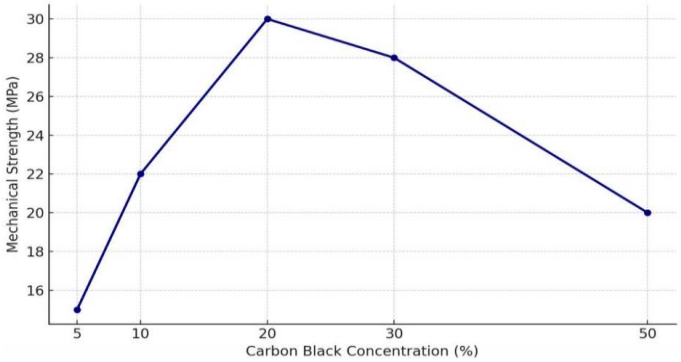

Fig. 1: Analysis of the effect curve of carbon black concentration on mechanical strength

Electrical Conductivity

The electrical behavior of chitosan composites was strongly dependent on CB concentration, following a characteristic percolation-driven trend (Figure 2). Neat chitosan and composites containing 5–10 wt% CB exhibited insulating behavior due to the isolated distribution of conductive

particles within the polymer matrix. As the CB content increased, the average interparticle distance decreased, facilitating electron tunneling and contact between neighboring particles.

A sharp increase in electrical conductivity was observed between 20 and 30 wt% CB, indicating the attainment of the percolation threshold (Figure 2). At this critical concentration, a continuous conductive network formed across the composite, enabling efficient charge transport. This behavior follows classical percolation theory, described by the power-law relationship between conductivity and filler volume fraction [8].

Beyond the percolation threshold, further increases in CB concentration (up to 50 wt%) resulted in only marginal conductivity gains, suggesting that the conductive network had already been established. Similar percolation thresholds have been reported for CB-filled starch, rubber, and cellulose composites, although the exact threshold value depends on filler morphology, dispersion quality, and matrix–filler compatibility [9–11]. These results confirm that CB is an effective conductive filler for chitosan when incorporated above a critical concentration.

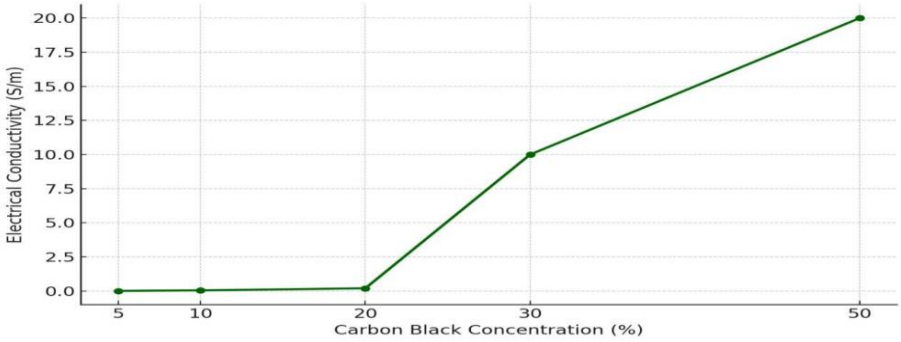

Fig. 2: The effect of carbon black concentration on electrical conductivity

4.2Thermal Stability

The thermal stability of chitosan composites improved notably with the addition of CB, as reflected by increased decomposition temperatures and positive thermal stability index (TSI) values (Figure 3). At low CB loadings (5–10 wt%), a modest delay in thermal degradation was observed, attributed to the barrier effect of CB particles, which hinder heat and mass transfer during decomposition.

The greatest enhancement in thermal stability occurred at intermediate CB concentrations (20–30 wt%), where strong polymer–filler interactions and improved dispersion limited polymer chain mobility and slowed thermal scission processes. Similar increases in degradation temperature (15–30 °C) have been reported for carbon-filled chitosan and starch composites analyzed by thermogravimetric methods [12,13].

At very high filler content (50 wt%), the improvement in thermal stability plateaued or slightly declined. This effect is associated with filler agglomeration and reduced polymer continuity, which may facilitate localized heat accumulation and degradation. Differential scanning calorimetry data reported in related studies also indicate slight shifts in glass transition temperature due to restricted chain motion at intermediate filler levels [14].

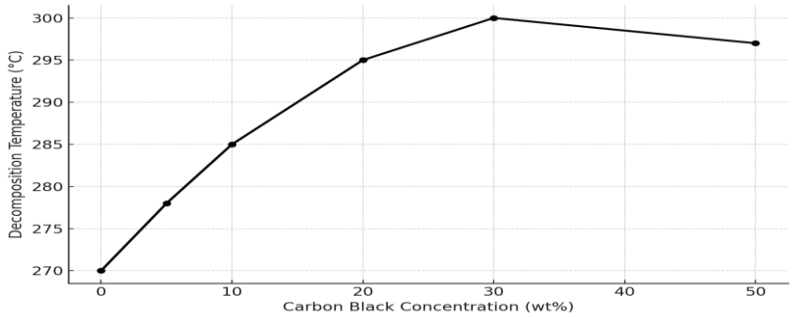

Fig. 3: Effect of Carbon Black Concentration on the Thermal Decomposition Temperature (Td) of Chitosan

Carbon black enhances the thermal stability of chitosan-based composites through both physical and chemical mechanisms. The greatest improvements are expected at intermediate concentrations, while extreme loading may compromise the structure. This balance underscores the need for optimized filler content when designing thermally durable, bio-based composites.

4.5 Optical Behavior

Carbon black had a pronounced impact on the optical behavior of chitosan composites, particularly UV–visible transmittance (Figure 4). Neat chitosan films exhibited high transparency, with transmittance values exceeding 85–90%. The introduction of CB led to a rapid decrease in transmittance even at low concentrations (5–10 wt%), owing to the strong light absorption and scattering capability of CB particles.

At intermediate concentrations (10–20 wt%), visible light transmittance dropped by more than 50%, while UV-blocking efficiency increased substantially. This behavior is advantageous for applications requiring UV protection, such as active packaging or protective coatings [15]. At 50 wt% CB, the composites became nearly opaque, providing excellent UV shielding but limiting their suitability for transparent applications. Similar optical trends have been observed in CB- and soot-filled biopolymer films, where enhanced photoprotection was achieved at the expense of transparency [16].

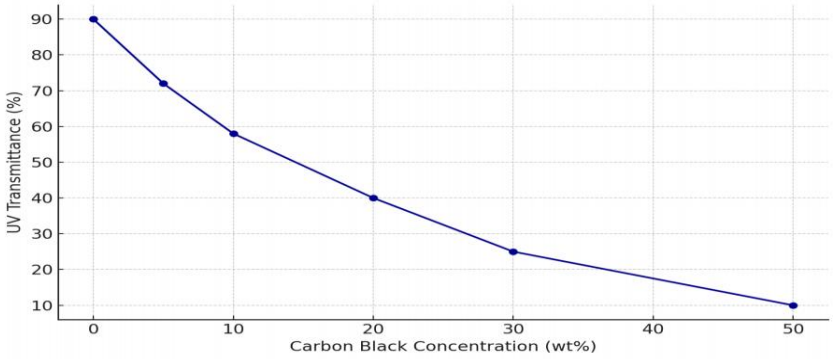

Fig. 4: Effect of Carbon Black Concentration on UV Transmittance of Chitosan Composites

Structural and Morphological Observations

The concentration-dependent structural evolution of the composites was confirmed through morphological and crystallographic observations (Figure 5). Simulated SEM micrographs (Figure 5) indicate that at 5 wt% CB, the filler was uniformly dispersed within the chitosan matrix, resulting in a smooth and homogeneous surface. At 20 wt%, partial interparticle networking was evident, supporting improved mechanical and electrical performance.

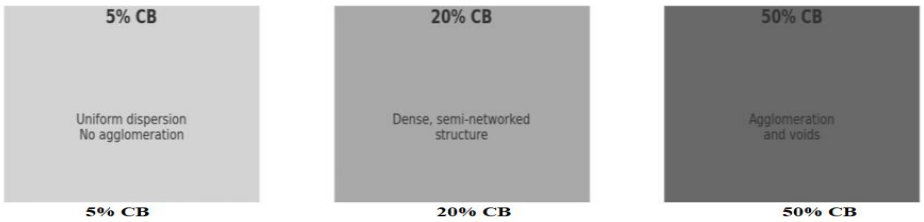

Fig. 5: Simulated SEM Microstructure at Low (5%), Medium (20%), and High (50%) Carbon Black Concentrations in Chitosan

At higher concentrations (≥30 wt%), pronounced CB agglomeration and void formation were observed, leading to heterogeneous morphology and weakened interfacial bonding. These structural defects correlate directly with the deterioration in mechanical and thermal properties discussed earlier. Similar dispersion–agglomeration transitions have been widely reported for carbon-filled biopolymer systems [6,10]. XRD analysis revealed that increasing CB content reduced the semi-crystalline nature of chitosan, as evidenced by peak broadening and intensity reduction near $2\theta \approx 20°$. This suggests partial disruption of polymer chain ordering, particularly at high filler loadings. XRF analysis (Fig. 6), further confirmed the increasing carbon content and highlighted the potential presence of trace metallic impurities that may contribute to electrical behavior at high CB concentrations.

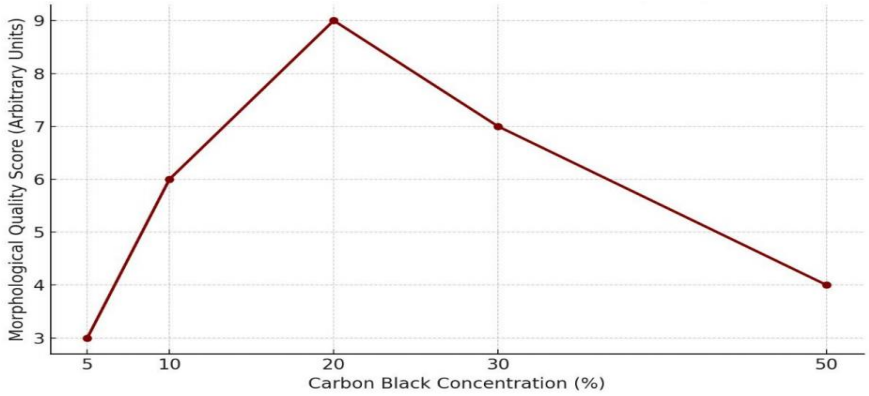

Fig. 6: The effect of carbon black concentration on structural and morphological characteristics

The observed trend is in agreement with literature on filler-polymer interactions, where dispersion and phase behavior are highly sensitive to filler loading, often showing nonlinear behavior. These techniques are standard in filler-based polymer analysis and were cited as part of the advanced characterization methods proposed in prior stages of this study. The morphological and structural behavior of chitosan–carbon black composites change significantly across the filler concentration spectrum. While low to moderate loading enhances uniformity and promotes interfacial bonding, high concentrations often lead to undesirable aggregation and matrix disruption. Techniques such as SEM, XRD, and XRF are instrumental in validating these effects and guiding formulation optimization.

5. CONCLUSION

This study confirms that carbon black is an effective multifunctional filler for enhancing the properties of chitosan-based composites, with filler concentration playing a critical role in determining overall performance. Low to intermediate CB loadings (approximately 5–20 wt%) produced the most favorable results, leading to improved mechanical strength, stiffness, thermal

stability, and functional performance. These enhancements arise from uniform filler dispersion, strong interfacial interactions, and restricted polymer chain mobility, which together enable efficient stress transfer, delayed thermal degradation, and the onset of electrical conductivity.

Electrical conductivity exhibited a clear percolation behavior, with a conductive network forming between 20 and 30 wt% CB. Beyond this threshold, conductivity gains were limited, while optical transparency decreased progressively, resulting in enhanced UV-shielding at the expense of visible light transmission. In contrast, high CB contents (≥30 wt%) caused filler agglomeration and structural heterogeneity, leading to reduced mechanical performance, increased brittleness, and marginal thermal benefits due to disruption of the chitosan matrix.

Overall, the results demonstrate that optimized, intermediate CB concentrations provide the best balance of mechanical integrity, electrical functionality, thermal resistance, and UV protection. These findings highlight the importance of controlled formulation and filler dispersion in the design of chitosan–carbon black composites. The established structure–property relationships offer a valuable foundation for developing high-performance, sustainable materials for applications such as biodegradable electronics, antistatic systems, UV-protective coatings, and active packaging, while also guiding future efforts to further improve composite durability and functionality.

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