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Formulation and Features of Chitosan and Natural Fiber Blended Bio-composite towards Environmental Sustainability

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ABSTRACT

This experimental investigation deals with the mechanical and tribological characteristics of biocomposite material made from polylactic acid (PLA), kenaf fiber (KF) and nano chitosan particles (nCp) through injection moulding approach for possible applications in biodegradable packaging, tissue engineering, wound dressing, and interior components of automobiles. Varying proportions of nCp (extracted from the crab shell) viz., 0, 1, 2, 3, and 4 wt.% are added in PLA matrix, strengthened with 30% of KF. The prepared composites are characterized as per the ASTM standard. Research indicates that incorporating 3 wt.% nCp into PLA-KF composites, produces noticeable improvements in mechanical strength. nCp acts as an effective interfacial agent, improving the bonding between PLA and KF leading to better stress distribution and higher mechanical performance. However, higher concentrations of nCp result in diminishing properties due to agglomeration or phase separation. nCp acts as a filler or surface modifier that enhances the composite's ability to withstand wear and reduce friction between surfaces in contact. Higher nCp addition (4 wt.%) results in poor dispersion, resulting in uneven stress distribution and weaker bonding of fiber with matrix, surface defects and accelerated material degradation occurs during use.

Keywords: PLA; Kenaf fiber; Chitosan; Injection moulding; Tensile Strength; Tribological properties.

1. INTRODUCTION

Biodegradable polymer composites reinforced with natural fibers are an emerging class of sustainable materials designed to reduce environmental impact while providing functional properties suitable for various applications. These composites are typically made from decomposable polymers viz., polyhydroxyalkanoates (PHA), poly lactic acid (PLA), or starch-based plastics, combined with natural fibers such as flax, kenaf, hemp, jute, or sisal. The natural fibers serve as reinforcement, refining the mechanical strength, durability, and toughness of the polymer matrix while maintaining biodegradability. These resources bid several advantages, including a lower carbon footprint, reduced necessity on petroleum-based plastics, and the ability to decompose naturally, minimizing long-term waste. Biodegradable polymer composites with natural fibers are increasingly used in packaging, automotive components, construction, and biomedical devices, as they align with the growing demand for eco-friendly, sustainable alternatives in material science and manufacturing.

Biodegradable polymer composites reinforced with chitosan derived from crab shells represent an innovative and eco-friendly material solution with significant potential across various industries. Chitosan, a biopolymer extracted from chitin found in crab shells, offers unique features like biodegradability, antimicrobial activity, biocompatibility, and good mechanical strength. When combined with biodegradable PLA, polycaprolactone (PCL), or starch-based plastics, chitosan enhances the composite’s strength and barrier properties while maintaining its environmentally friendly nature. These composites are especially valuable in medicinal uses in the fields of wound care, tissue engineering, and drug administration, as well as in packaging and agriculture, where their antimicrobial properties help extend product shelf life and reduce contamination. The use of chitosan from crab shells also supports waste valorization, as it transforms crustacean shell waste into high-value materials, contributing to sustainability and circular economy initiatives.

Composites using kenaf fibers (KFs) and a PLA resin of the emulsion type were created to be biodegradable in both directions. The flexural strength of the unidirectional fiber-reinforced composites was 254 MPa and the tensile strength was 223 MPa; both values grew proportionally up to a fiber proportion of 50%. Composting kenaf/PLA composites for a period of four weeks causes a 91% drop in tensile strength and a 38% drop in weight as the composites decompose (Ochi *et al.* 2008). The kenaf/PLA composites' mechanical as well as thermal characteristics were found to be improved by chemical changes. An increase in the kenaf loading fraction is associated with improved degradation of the composites (Khan *et al.* 2023). Various fiber ratios were experimentally studied to determine their effect on both the thermal and mechanical attributes of PLA-KF composites. Woven KF has enhanced the composites' flexural, impact, and tensile strengths. With a tensile strength of 61 MPa, flexural strength of 62 MPa, and impact strength of 48 kJ/m^2, the WK40 produced the highest results. However, after adding 40% WKF, the composites' flexural modulus was marginally lowered by 6.7%. Thermogravimetric testing revealed that the WK40 sample produced the most residue and improved the heat resistance and stability of the composites (Nor *et al.* 2022). After the fibers were chemically treated, PLA-KF composites were manufactured via melt compounding and compression molding. The flexural strength was significantly improved by around 27% at a concentration of 1.0M thanks to the fiber surface treatment. Compared to their untreated counterparts, treated KBC and KCC exhibited better impact characteristics. According to (Surip *et al.* 2017), the flexural strength of KCC1.0 was 43.4% higher than that of KCC0.5.

Kumar and Srinivasan reinforced chitosan particles (Cp) in basalt fiber reinforce PLA matrix using injection moulding. The proportion of crystallinity and the glass transition temperature both rise with increasing amounts of fiber and filler. To improve the composites' physical and thermal stability, the ideal weight proportion of basalt fiber was determined to be 25% and that of chitosan to be 10% (Arul and Srinivasan, 2016). Researchers used compression molding and melted mixing to create PLA-Cp composite. Tensile testing on dried samples revealed a 2% increase in tensile strength and a 14% increase in tensile modulus compared to pure PLA, which had 2.5 php of Cp added to it. The thermal study revealed that neat PLA's crystallinity proportion was 51% higher with a chitosan loading rise of up to 10 php, whereas water intake increases with Cp loading (Kamaludin *et al.* 2021). Development of PLA-Cp scaffolds using fused filament fabrication (FFF) was done. Because the inclusion of Cp introduces abnormalities in the polymeric chains, the tensile strength falls as the weight portion of chitosan rises. As the infill density increased, TS was able to rise. Since the composite material achieves a higher density as the Cp fraction grows, the compressive strength also increases. Similar to tensile, flexural strength exhibits a mix of brittle and ductile ruptures because the material is subject to both tensile and compressive pressures simultaneously (Singh *et al.* 2020). Biocomposite of PLA/cellulose nanofibers (CNF) improved by the using amphiphilic chitosan synthesized by substitution process. A biocomposite of PLA, chitosan, and CNF demonstrated improved mechanical, thermal, and miscibility characteristics without agglomeration. Compared to clean PLA, the composite exhibited less hydrophobicity in the wettability test (Nasution *et al.* 2021).

The formulation of chitosan and natural fiber-blended bio-composites introduces several innovative aspects that significantly advance environmental sustainability. Chitosan, derived from chitin found in crustacean shells, is biodegradable, renewable, and exhibits natural antimicrobial properties, making it an eco-friendly alternative to petroleum-based polymers. When combined with natural fibers (kenaf, vetiver, aloe vera, hemp, flax, or jute), the composite gains strength, flexibility, and thermal stability, which enhances its suitability for a wide range of applications, from packaging to automotive parts. This blend reduces reliance on synthetic materials and curbs plastic pollution, as it breaks down naturally in the environment, mitigating waste accumulation. Additionally, the sourcing of both chitosan and natural fibers is generally low-energy and generates fewer greenhouse gas emissions compared to traditional plastic production. Thus, these bio-composites provide a sustainable alternative with a reduced environmental footprint, contributing to the circular economy by enabling materials that are renewable, biodegradable, and potentially recyclable.

From the literature, it was found that no studies were performed on the combination of PLA-KF-Cp subjected to mechanical and wear analysis. The incorporation of nano Cp (nCp) can enhance the mechanical strength of the biodegradable nanopolymer composites along with improved thermal stability. Hence, this study's originality lies in performing mechanical tests on the newly developed PLA-KF-nCp composite from injection moulding following the guidelines set forth by the ASTM. Also, wear analysis was done on the fabricated composite to understand its mechanism of wear for suitability in lightweight automotive applications.

2. MATERIALS AND METHODS

PLA is a biodegradable and bio-based polymer known for its favorable physical and mechanical properties and its wide range of applications. Physically, PLA is a thermoplastic with a density of about 1.25 g/cm³, a melting point ranging from 160°C, and a glass transition temperature of 55 °C. PLA has good tensile strength (65 MPa) and modulus (3 GPa) and is widely used in packaging (films, bottles, containers), biomedical devices (sutures, implants, drug delivery systems), 3D printing filaments, and disposable products like utensils and bags. Its biodegradability makes it a sustainable alternative to petroleum-based plastics, especially in industries focused on reducing environmental impact (Ranakoti *et al.* 2022).

Kenaf fiber (KF) is a natural plant-based fiber that is lightweight, with a density ranging from 1.3 g/cm³. They have a high cellulose content (approximately 65–75%), contributing to their strength and durability (Rahman *et al.* 2024). The fibers are relatively long, with lengths up to 2.5 meters, and exhibit good moisture absorption. KFs exhibit impressive tensile strength (270–930 MPa) and Young's modulus of around 14–53 GPa, which makes them comparable to traditional synthetic fibers like glass fibers. KFs are commonly used in automotive components (e.g., door panels, dashboards) due to their lightweight and high strength, which help improve fuel efficiency (Bhambure *et al.* 2023).

A natural biopolymer known as chitosan is produced by deacetylating chitin, an ingredient present in crustacean shells such as crabs. Because of its non-toxicity, biocompatibility, and biodegradability, chitosan finds extensive use in the fields of medicine, pharmaceuticals, and ecology (Hisham *et al.* 2024). It also exhibits antimicrobial and antifungal properties. Chitosan exhibits moderate tensile strength (20–50 MPa), depending on its molecular weight and form, and a Young's modulus in the range of 0.5–2 GPa, making it strong yet flexible (Alemu *et al.* 2023). The density of nano chitosan nanoparticle (nCp) is 0.3 g/cm^3, procured from Intelligent Materials Private Limited, India.

Fig. 1: (a) PLA pellets (b) Raw KF and (c) Chitosan nanopowder

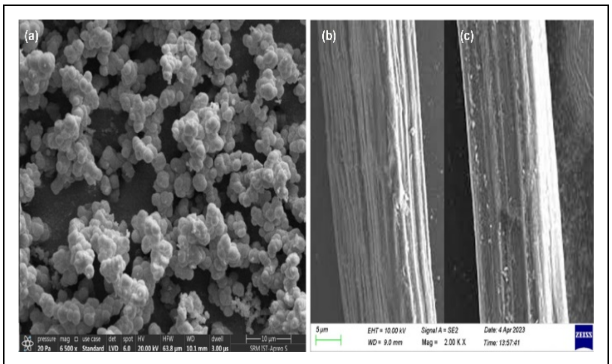

Fig. 2: Chitosan nanoparticle SEM and KFs before and after NaOH treatment

Figure 1 presents the PLA pellets procured from Natur Tec India Pvt Ltd, Chennai, KF from Go Green Products, Chennai and nCp from Intelligent Materials Private Limited, Punjab, India. The KFs (30 wt.%) were cut into smaller pieces and is mixed well with the PLA pellets along with the required quantify of nCp (0, 1, 2, 3, and 4 wt.%) termed as PKC0, PKC1, PKC2, PKC3, and PKC4 and blended in a twin-screw extruder, which is then sent to the injection moulding machine, set at a temperature of 180 °C, for complete melting and flow of PLA pellets for better moulding behaviour (Vasanthkumar *et al.* 2022). The moulded part is then ejected from the die after cooling. Figure 2 presents the SEM image of the nCp and the KF before and after chemical treatment. The 6 wt.% of NaOH treatment on KF eliminates impurities such as lignin, hemicellulose, and waxes on the surface of KFs, resulting in a cleaner and rougher texture. This roughness improves the fiber's interfacial bonding with matrices in composite materials (Kumar *et al.* 2022).

Blending chitosan with natural KFs significantly enhances both the mechanical and biodegradable properties of bio-composites, making them superior to many traditional materials. Chitosan contributes to the bio-composite's structural integrity and antimicrobial qualities, while KFs, known for their high tensile strength and low density, reinforce the composite matrix, resulting in increased durability, flexibility, and impact resistance. This combination offers mechanical strength comparable to synthetic composites but with the added advantage of being lighter and biodegradable. KFs, derived from renewable plant source, also improve water resistance and overall stability of the composite, complementing chitosan's natural degradability. Together, these materials decompose more efficiently under natural environmental conditions, thus reducing long-term waste and contributing to a sustainable lifecycle. This synergy between chitosan and KFs provides a renewable alternative that lessens environmental impact, addressing both mechanical performance and biodegradability in a way that traditional, petroleum-based materials cannot.

3. RESULTS AND DISCUSSIONS

After fabricating the composite, scanning electron microscope (SEM) images were captured to identify the distribution of KFs and nCp in the matrix of PLA. Figure 3 presents the SEM of the composite. In the preparation of a biocomposite material using injection molding, KFs and nCp are uniformly distributed within a PLA which enhances the mechanical and thermal properties. The uniform dispersion ensures that the material's stress is dispersed uniformly, which increases its overall strength and decreases the probability of weak spots, stiffness, and durability of the composite (Han *et al.* 2012). The injection molding process, with its precise control over temperature and pressure, facilitates the even distribution of KFs and nCp within the PLA matrix, resulting in a high-quality biocomposite (Rabbi *et al.* 2021).

Chitosan-KF bio-composites possess unique functional properties, including inherent antimicrobial activity and enhanced water resistance, which set them apart from other biodegradable materials. Chitosan naturally exhibits strong antimicrobial properties against a range of bacteria and fungi, making the composite ideal for applications where hygiene is essential, such as in packaging, medical devices, or food storage. This antimicrobial quality is a significant advantage over other biodegradable materials that lack this functionality, reducing the need for additional chemical treatments. Additionally, the inclusion of KFs improves the bio-composite's structural stability and water resistance, helping it withstand moisture and mechanical stress better than many other bio-based composites. This water-resistant capability stems from KFs natural hydrophobicity and reinforces the composite's resilience in humid environments. Together, these properties create a durable, hygienic, and sustainable material that is especially useful in industries seeking green alternatives without compromising on functional performance, distinguishing chitosan-KF bio-composites in the landscape of biodegradable materials.

Adding nCp up to 3% in a PLA matrix reinforced with 30% KF improves the tensile strength through enhanced bonding at interfaces of PLA and KFs, and nCp as seen in Figure 4. At this optimal concentration, the nCp effectively fills the gaps between the PLA and KF surfaces, improving stress transfer and reinforcing the composite. However, beyond 3%, a decline in tensile strength is observed, likely due to agglomeration of the excess nCp. When nCp clusters, they create localized stress points and hinder the even distribution of load within the matrix (Torres *et al.* 2018). This disrupts the polymer-fiber bonding, resulting in poor mechanical performance and decreased tensile strength. Thus, excessive nanoparticle loading reduces the composite's structural integrity instead of enhancing it. The KFs, with their high strength and stiffness, enhance the tensile properties by effectively transferring the load from the weaker PLA matrix to the stronger fibers, allowing the composite to withstand higher tensile forces before breaking (Ochi *et al.* 2008; Gunasekar *et al.* 2024). When compared with PKC0, the PKC3 had an improved tensile strength by 18.65%, but comparing the PKC3 with PKC4, the tensile strength decreased by 2.31%.

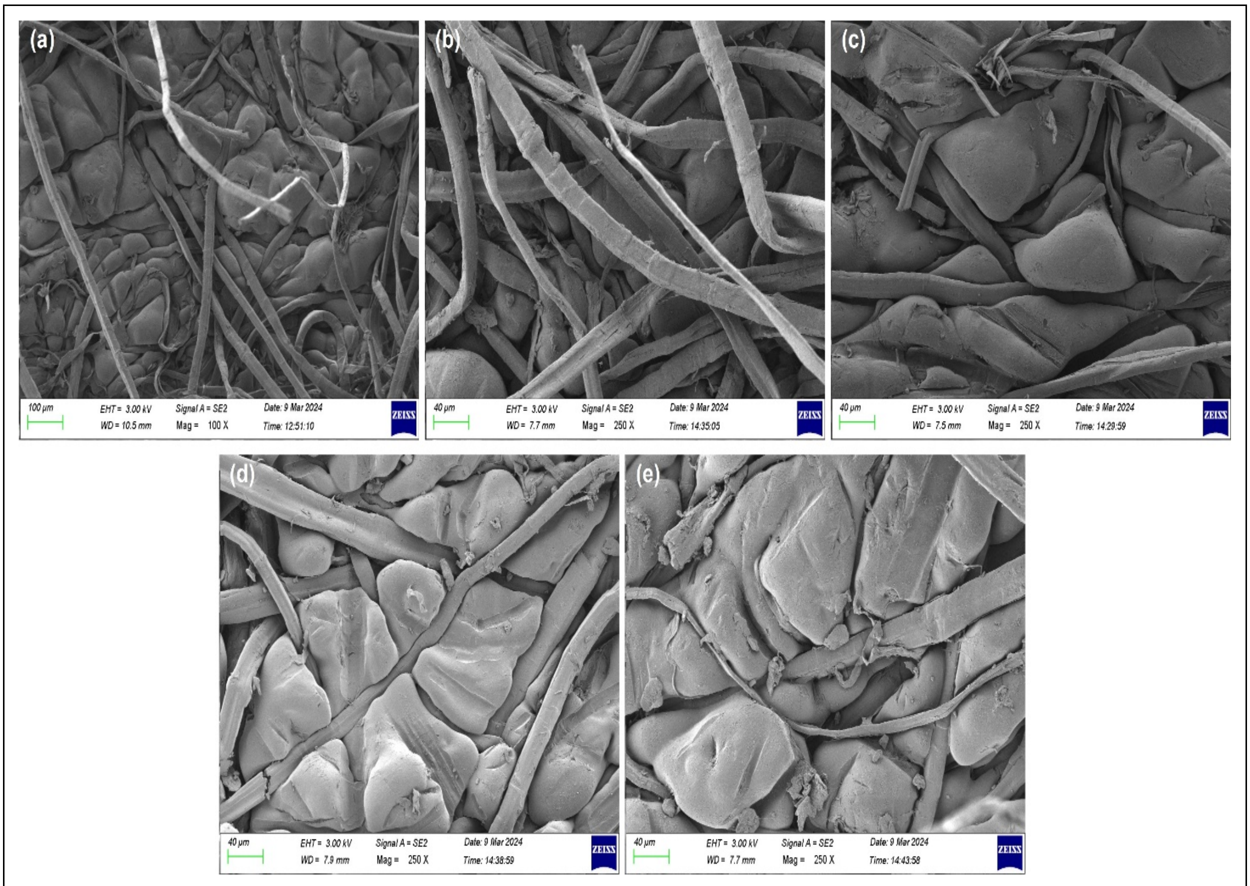

Fig. 3: SEM of fabricated BPNC (a) PKC0 (b) PKC1 (c) PKC2 (d) PKC3 (e) PKC4

The fractured surface of PLA reinforced with 30 wt.% KFs and different proportions of nCp after a tensile test typically exhibits a rough and fibrous appearance, as seen in Figure 5. The fracture surface reveals regions where the KFs have pulled out, indicating poor fiber-matrix adhesion, and areas where fibers have fractured, showing that some fibers carried substantial loads before failure (Suzuki *et al.* 2013). The matrix material (PLA) surrounding the fibers often shows signs of plastic deformation, such as crazing or micro-cracks. In some cases, voids or gaps between the fibers and the matrix may be visible, suggesting debonding or insufficient wetting in the course of making composites. Overall, the fracture pattern is a mix of brittle fracture in the PLA and fiber pull-out or breakage, with the failure mode reflecting the balance between the matrix's brittleness and the reinforcing role of the KFs (Singh *et al.* 2024).

Figure 6 presents the impact strength of the fabricated BPNC, the strength increases until 3% addition of nCp, after which a decreasing trend is seen as depicted. The PLA+30%KF+3%nCp enhances the impact strength due to the improved energy absorption and distribution capabilities of the composite (Sathish *et al.* 2024). The reinforced nCp acts as a toughening agent, filling micro-voids and improving the interaction among KFs and PLA, resulting in better resistance to sudden impacts (Zakaria *et al.* 2013). The nCp helps dissipate the impact energy more effectively, reducing the likelihood of crack initiation and propagation. However, beyond the 3% threshold, a decreasing trend in strength is seen due to the nCp agglomeration, hindering the uniform load distribution and weakening the interfacial strength among PLA and KFs. The reinforced KFs absorb and dissipate energy during impact, reducing the brittleness of PLA and increasing the material's toughness (Özturk *et al.* 2010; Tharazi *et al.* 2023). The PKC3 produced a maximum impact strength of 45.85 kJ/m^2, which is 26.34% higher than the PKC0 composite strength. With PKC4, the impact strength is lowered by 8.05% as compared with PKC3 composite.

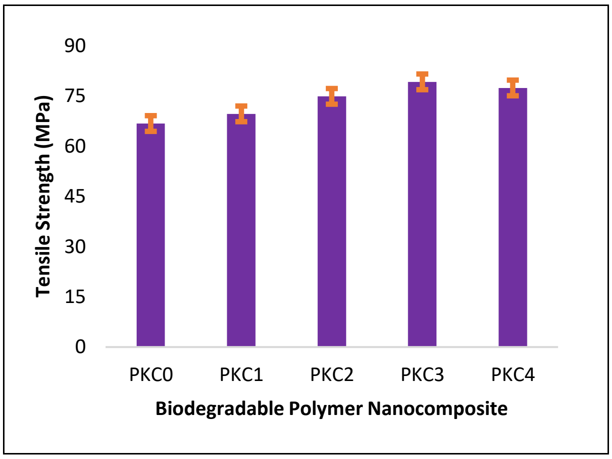

Fig. 4: Tensile strength of BPNCs

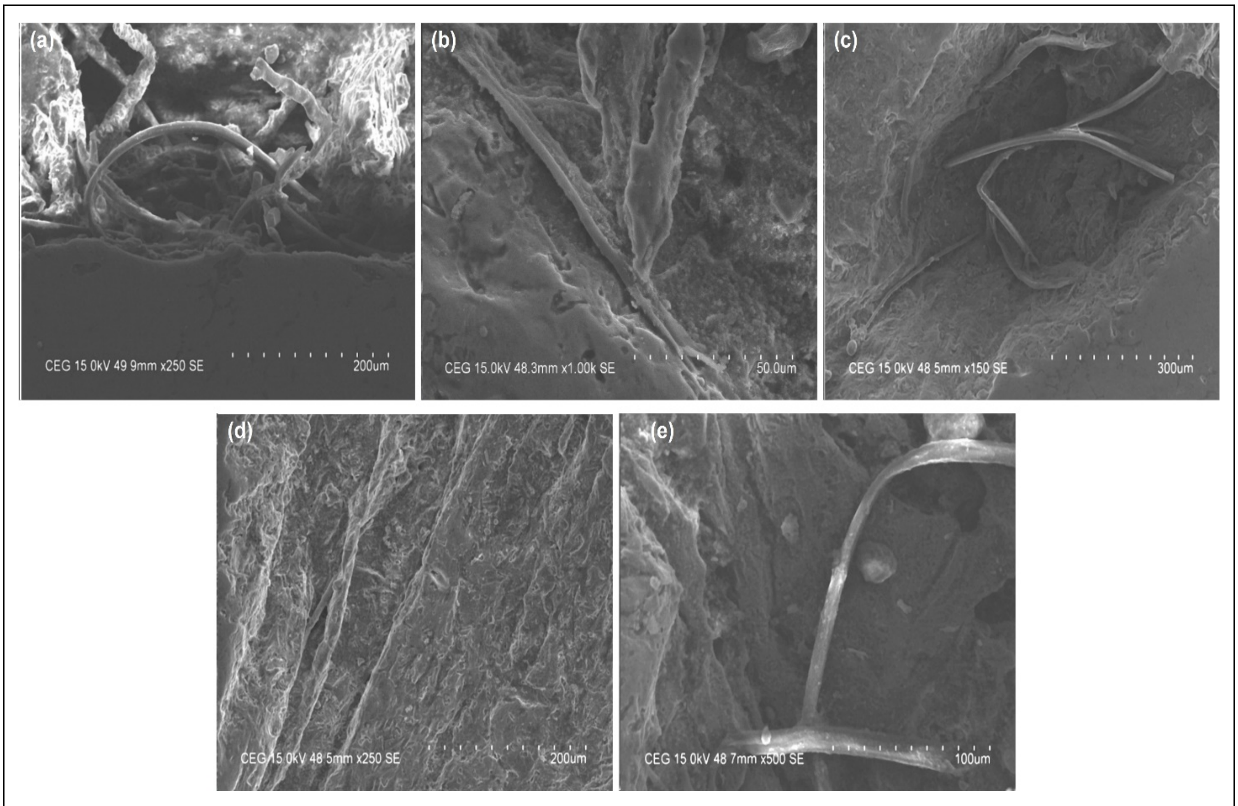

Fig. 5: Fractured surface of BPNC (a) PKC0 (b) PKC1 (c) PKC2 (d) PKC3 and (e) PKC4

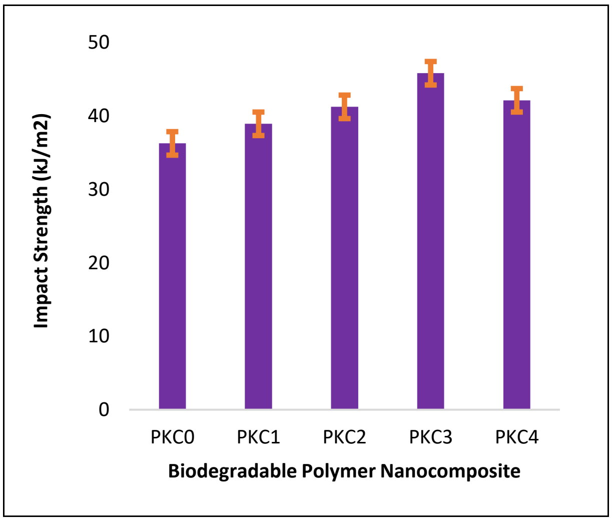

Fig. 6: Impact strength of BPNCs

The flexural strength of BPNCs evaluated is shown in Figure 7. The addition of nCp up to 3% in a PLA+30%KF composite enhances the flexural strength because of higher interfacial bonding and better stress transfer among KFs to PLA. The nCp fills voids and enhances the adhesion between PLA and the KFs, resulting in a stiffer composite that can resist bending forces more effectively. At this optimal concentration, the composite's ability to bear and distribute flexural loads improves, leading to higher flexural strength (Rajamuneeswaran *et al.* 2015). However, beyond 3%, flexural strength lowers as the tendency of nCp to agglomerate, creating localized stress points and disrupting the uniform distribution of load across the matrix (Li *et al.* 2004). This results in a reduction in tensile strength, as the composite becomes more prone to failure under tensile stress despite the initial improvement in flexural strength.

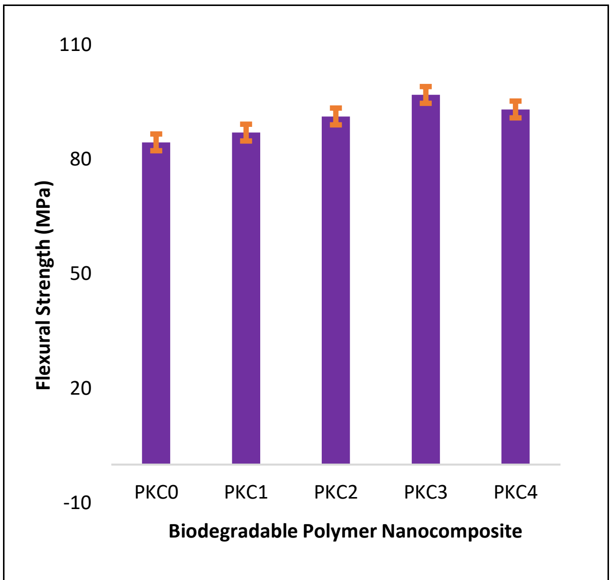

Fig. 7: Flexural strength of BPNCs

Additionally, the incorporation of KFs improves the flexural strength by providing structural reinforcement, allowing the composite to resist bending forces more effectively. The fibers act as bridges that restrict crack propagation and provide stiffness under flexural loads, resulting in a composite that is stronger and more resilient across multiple mechanical properties (Atiqah *et al.* 2024). With a flexural strength of 96.85 MPa, the PKC3 sample had an increased flexural strength by 14.72, as compared with PKC0 composite, whereas with further addition of nCp (PKC4) a 3.93% reduction in flexural strength is seen.

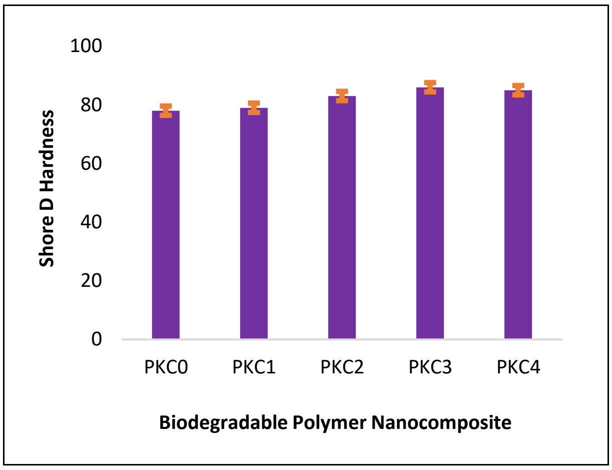

Fig. 8: Hardness of BPNCs

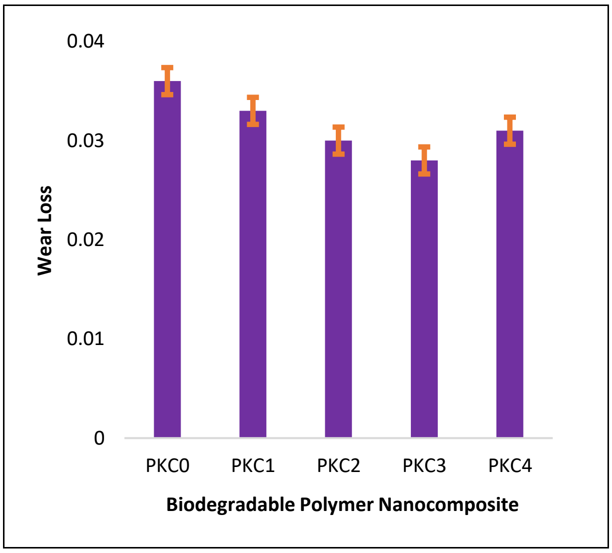

Fig. 9: Wear loss of BPNCs

Figure 8 presents the shore D hardness obtained for different BPNCs. The addition of 3 wt.% nCp in PLA+30%KF matrix increases the Shore D hardness due to the enhanced stiffness and rigidity of the composite. An improved interaction among PLA and KFs, along with the filling of micro-voids inside the matrix by the nCp, leads to a denser and better compact structure. This increased structural integrity enhances the composite's resistance to surface indentation, reflected in the improved Shore D hardness (Hui *et al.* 2023). However, beyond 3 wt.% of nCp, a decrease in Shore D hardness is observed due to clustering and agglomeration of nCp. This decline is attributed to the agglomeration of excess nano chitosan particles. These agglomerates create weak points in the composite structure, reducing the overall density and uniformity. As a result, the composite becomes less rigid and more prone to surface deformation, leading to a reduction in Shore D hardness (Senthilkumar *et al.* 2024). The shore D hardness obtained by the PKC3 composite is 10.26% higher than the PKC0 counterpart, whereas a decrease in hardness is noticed with PKC4 composite (1.16%).

The nCp acts as a lubricant and reinforces the composite's structure by filling voids and strengthening the adhesion between the PLA and KFs thereby lowering the wear loss of BPNCs (Kumar *et al.* 2020) as depicted in Figure 9. This leads to a more uniform and compact surface, which resists abrasion and wear more effectively, reducing material loss during frictional contact (Navarro *et al.* 2012). As more nCp (4 wt.%) are added, they tend to cluster, leading to poor dispersion and creating weak zones in the composite structure. These agglomerates can act as stress concentrators, promoting uneven wear and increase the roughness of the surface. This undermines the integrity of the composite and makes it more susceptible to material degradation during friction, ultimately resulting in higher wear loss (Arul *et al.* 2018). The wear resistivity of PKC3 is higher than all the other combinations, producing a wear loss of 0.028g, which is 22.22% lower than the PKC0 sample. But higher addition of nCp (PKC4) there is an increase in wear loss by 10.71% due to poor bonding.

Formulating chitosan-KF bio-composites through injection molding presented several technical challenges, primarily due to the differences in thermal stability and flow properties between chitosan and natural fibers like kenaf. Chitosan's sensitivity to high temperatures made it prone to degradation during the molding process, which typically requires sustained high heat to ensure proper flow and form within the mould. To address this, processing conditions were carefully optimized, including adjusting the injection temperature and pressure to prevent chitosan from degrading while still achieving effective molding. Additionally, KFs, being prone to uneven dispersion under pressure, required specific modifications to ensure a uniform blend; this included surface treatment of the fibers to improve their compatibility with the chitosan matrix and incorporating low-impact additives to enhance flow without compromising biodegradability. These adjustments allowed the composite to maintain structural integrity and biodegrade efficiently, supporting environmental sustainability by reducing the reliance on petroleum-based polymers and enabling production of a consistent, renewable bio-composite material suitable for various applications.

Incorporating 3% chitosan into PLA-KF composites optimizes the mechanical performance by enhancing both the strength and flexibility of the composite matrix. At this concentration, chitosan effectively strengthens the bonding interface between PLA and KFs, improving stress transfer and creating a more integrated and resilient composite structure. Higher or lower amounts of chitosan tend to disrupt this balance: too much chitosan can lead to agglomeration, causing weak points and reducing uniformity, while too little chitosan does not sufficiently improve fiber-matrix adhesion. The 3% chitosan level achieves an ideal synergy, where the material gains toughness without compromising PLA's biodegradable properties. This composition enhances the tensile strength, impact resistance, and ductility of the composite, making it better suited for structural applications while maintaining environmental sustainability. This precise balance between performance and eco-friendliness makes the 3% chitosan content particularly advantageous for high-performance, biodegradable composites.

4. CONCLUSION

The injection moulded PLA+30%KF+x%nCp samples containing alkali treated KFs were subjected to mechanical and wear tests and the findings obtained are as follows.

- SEM images show that KFs and nCp are uniformly distributed within a PLA, which enhances the mechanical and thermal properties as stresses will be evenly distributed throughout the material, reducing the weak points and enhancing the durability and strength of the composite.

- Increased interfacial bonding among the KFs and 3 wt.% of nCp in the PLA matrix increases the tensile, impact, and flexural strength of the composite as the nCp effectively fills the gaps between the PLA and KF surfaces, improving stress transfer and reinforcing the composite.

- A higher addition of nCp (PKC4) shows a decrease in mechanical and wear resistance due to the clustering of nCp paving the way for lower interfacial bonding and strength.

- The fracture surface reveals regions where the KFs have pulled out, indicating poor fiber-matrix adhesion and that some KFs carried substantial loads before failure. The PLA surrounding the fibers shows signs of plastic deformation and micro-cracks.

- The self-lubricating characteristics of nCp lowers the wear resistance of the BPNCs, but higher addition (PKC4) leads to poor wear behaviour due to clustering of nCp and poor bonding.

- The tensile, impact, flexural and shore D hardness of PKC3 is increased by 18.65, 26.34, 14.72, and 10.26% when compared with PKC0. But the wear loss is lowered by 22.22%, showing higher durability. With PKC4, the properties tend to lower.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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