Morphological and mechanical properties of chitosan/cellulose nanofibrils/aspirin polymer nanocomposite films

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Abstract

This study examined the mechanical properties of composite films composed of chitosan, cellulose nanofibrils, and aspirin. This biomaterial has promising characteristics and holds potential for various applications. The composite material, which was synthesised using precise fabrication techniques, consists of chitosan as a biocompatible substrate, cellulose nanofibrils for enhancing structural integrity, and aspirin for additional therapeutic benefits. The composite material exhibited increased tensile strength, tensile modulus, and elongation at break. The experimental results demonstrate that the tensile strength and tensile modulus exhibit an upward trend as the loading of cellulose nanofibrils (CNFs) increases. This observation suggests a synergistic improvement in mechanical robustness, which can be attributed to the combined effects of chitosan and CNFs. A reduction in elongation at break was seen as the loading of CNFs increased. The adaptability of the material is further emphasised by its tensile modulus and elongation at break. This study presents opportunities for the development of sustainable packaging materials, as the biodegradable properties of chitosan and cellulose are in line with

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current environmental priorities. The composite films composed of chitosan, cellulose nanofibrils, and aspirin demonstrate a notable combination of mechanical robustness and therapeutic properties. This research establishes the foundation for future attempts in biomaterial design by introducing a versatile composite that has the potential to significantly influence various sectors, including materials science and healthcare.

1. Introduction

The integration of biopolymers has become an intriguing pathway in the exploration of sustainable and versatile materials. The amalgamation of chitosan, cellulose nanofibrils (CNFs), and aspirin within composite films signifies a ground-breaking endeavour to merge the mechanical resilience of organic polymers with the therapeutic capabilities of a pharmaceutical substance [1]. This novel composite material exhibits potential for a wide range of applications, encompassing many fields such as biomedical engineering and sustainable packaging solutions. The primary constituent of this composite material, chitosan [2], is a biopolymer that is obtained from chitin, a widely distributed substance present in the exoskeletons of crustaceans [3]. Chitosan, known for its exceptional biocompatibility, serves as the fundamental component of the composite material. Its inclusion imparts mechanical strength and antibacterial characteristics, hence expanding its potential applications across several domains [4].

Cellulose nanofibrils, derived from cellulose, serve a crucial function in enhancing the mechanical strength of the composite material [5]. CNFs, or cellulose nanofibers, have been shown to possess a remarkable aspect ratio and intrinsic strength, as demonstrated by Vega-Vásquez *et al.* [6]. These properties make CNFs effective as reinforcing agents, hence enhancing their tensile strength and toughness. The structural matrix formed by the combination of chitosan and cellulose demonstrates remarkable mechanical performance, as reported by Wang *et al.* [7].

The incorporation of aspirin into the composite material introduces a medicinal aspect. According to Pawar *et al.* [8], aspirin is a widely recognised substance with proven anti-inflammatory and antiplatelet properties, hence contributing therapeutic qualities to the material. The combination of mechanical strength and therapeutic potential exhibited by this particular blend presents new opportunities for use in the biomedical domain, where materials must fulfill both mechanical requirements and contribute to the process of healing.

It is crucial to comprehend the complexities involved in the manufacturing process,

the arrangement of components at the nanoscale, and the resulting impact on its mechanical characteristics. The present study investigated the manufacturing procedures, conducted assessments of mechanical properties, and explored potential applications, thereby offering a full overview of the mechanical phenomena exhibited by composite films composed of chitosan, cellulose nanofibrils, and aspirin.

2. Materials and Methods

The Agave Sisalana plant, which was utilised in this research, was acquired from the Garkida Local Government Area in Adamawa State. The necessary analytical grade reagents were obtained from Marcy Surgical Limited in Gombe, Gombe state. These reagents include NaOH (97% purity), fused CaCl, H_2SO_4 (98% purity), H_2O_2 (50% purity), sodium sulphite, and NaCl (80% purity). Additionally, aspirin (acetylsalicylic acid), ibuprofen, diclofenac, and chitosan were procured from Cynco Chemicals and Co, Ltd in Lagos, Lagos state. The methodology employed for the extraction of cellulosic nanofibrils and the subsequent fabrication of nanocomposites was detailed in a study conducted by Sarkar *et al.*, [9]. The tensile properties of pure chitosan and cellulose nanofibrils nanocomposite films were evaluated utilising a universal testing equipment.

3. Results and Discussion

The physicochemical characterization of the composite films included analysis of their morphology and mechanical properties.

3.1. Scanning electron microscopy of chitosan-cellulose nanofibrils/aspirin nanocomposites



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Figure 1. Scanning electron microscopy (SEM) image of chitosan-cellulose nanofibrils/aspirin polymer nanocomposites.

The surface morphology of the initial materials plays a crucial role in the formulation of composites due to its influence on the swelling characteristics of the composite for drug delivery purposes, drug encapsulation, and release behaviour [10].

The scanning electron microscopy (SEM) pictures of the chitosan-cellulose nanofibrils/Aspirin nanocomposite films demonstrate a consistent and even surface, suggesting successful film production (see Figure 1). The presence of nanofibrils within the film matrix was also detected, indicating the successful integration of cellulose nanofibrils (see Figure 1(b and c)).

The scanning electron microscopy (SEM) image of the F0 Aspirin film (Figure 1(a)) exhibits a uniform surface morphology devoid of any discernible phase alterations, suggesting a homogeneous amalgamation of chitosan and aspirin. The F1 aspirin sample, which had a CNF level of 0.25%, demonstrated a uniform and dense dispersion of CNF inside the mix matrix, as depicted in Figure 1(b). The enhanced visibility of the uniform dispersion of cellulose nanofibers (CNF) on a rough surface can be attributed to their effective inclusion inside the chitosan matrix.

Likewise, the scanning electron microscopy (SEM) image indicated the presence of F4 Aspirin aggregation, as shown in Figure 1(c). The agglomeration phenomenon observed in composite systems can be attributed to the edge-edge interactions between hydroxyl groups present in the cellulose nanofibrils (CNF) [11]. The presence of uniformly embedded cellulose nanofibrils (CNF) in the film suggests a significant level of interaction between the CNF fillers and the matrix material. An increase in porosity

was detected when the content of cellulose nanofibers (CNF) in the polymer matrix increased. This suggests that CNF acts as a bridge between adjacent polymer chains, leading to enhanced porosity. The higher occurrence of CNF is observed in F1 and F4 as opposed to F0, which can be attributed to the lack of CNFs inside the chitosan matrix. The enhanced intermolecular interaction between the chitosan chains and CNFs resulted in the formation of a significantly more porous structure in comparison to the pure chitosan material.

Tensile Strength of Aspirin 70 57.8 60 53.5 50.9 50 43.7 36.8 40 (Mpa) 30 Aspirin 2010 0 F0F 1 F 2 F 3 F 4

3.2. Mechanical properties of chitosan-cellulose nanofibrils/aspirin polymer nanocomposites

Figure 2. Tensile strength of the cellulose polymer nanocomposite film containing aspirin (FA).



Figure 3. Tensile modulus of the cellulose polymer nanocomposite film containing aspirin (FA).





The consideration of mechanical qualities is crucial in the development of transdermal films intended for controlled release of medicines at specific disease target areas. In this study, we examined the tensile strength, tensile modulus, and percentage elongation at break of various formulations of aspirin nanocomposites, namely F0, F1, F2, F3 and F4.

Figure 1 displays the tensile strength of chitosan and CNF/chitosan films at varying levels of CNF loading. The findings indicate that the chitosan film (F0) exhibited a tensile strength of 36.8 MPa, a tensile modulus (E) of 105 MPa, and a percentage elongation at break (%EB) of 17.60%. The results clearly indicate that the incorporation of cellulose nanofibers (CNFs) in chitosan leads to an increase in its tensile strength. Specifically, the tensile strength values were observed to increase from 36.8 MPa to 43.7, 50.9, 53.5, and 57.8 MPa as the loading of CNFs (expressed as weight percent) rose from 0.25, 0.5, 0.75, to 1.0, respectively.

Figure 2 displays the tensile moduli (E) of chitosan and nanocomposites with different percentages of cellulose nanofibers (CNF). The tensile modulus of the chitosan film exhibited a rise when the loading of CNFs increased. Specifically, the tensile modulus values progressed from 105 MPa (F0) to 249.3 MPa (F1), 573.2 MPa (F2), 730.2 MPa (F3), and 920.2 MPa (F4). In contrast, the percentage elongation at break (%EB) of Chitosan film exhibited a decrease from 17.60% (F1) to 8.12% (F1), 5.30% (F2), 4.90% (F3), and 4.30% (F4), respectively, when the loading of CNFs increased, as depicted in Figure 3.

The aforementioned findings clearly indicate that when the proportion of CNFs in the chitosan matrix increases, there is a noticeable drop in elongation at break. Conversely, an increase in both tensile strength and tensile modulus is noted, which aligns with the typical behaviour reported in composite materials.

4. Conclusion

This study investigates the results of several studies, including tensile strength, flexural strength, hardness, and compressive strength, performed on composite films consisting of chitosan-cellulose nanofibrils and aspirin. The purpose of these analyses is to assess the potential of these films for drug loading applications. The findings of the study demonstrated that the inclusion of cellulose nanofibrils and aspirin within the chitosan matrix led to a substantial enhancement in the mechanical characteristics of the films. The composite films demonstrated improved tensile strength, flexural strength,

hardness, and compressive strength, rendering them appropriate for drug loading applications necessitating sturdy and long-lasting materials.

Conflict of Interest

No conflict of interest was declared.

References

- [1] Omidi, S., & Kakanejadifard, A. (2019). Modification of chitosan and chitosan nanoparticle by long chain pyridinium compounds: synthesis, characterisation, antibacterial, and antioxidant activities. *Carbohydrate Polymers*, 208, 477-485. <u>https://doi.org/10.1016/j.carbpol.2018.12.097</u>
- [2] Silva, M.M., Calado, R., Marto, J., Bettencourt, A., Almeida, A.J., & Gonçalves, L.M.D. (2017). Chitosan nanoparticles as a mucoadhesive drug delivery system for ocular administration. *Marine Drugs*, 15, 370. <u>https://doi.org/10.3390/md15120370</u>
- [3] Orasugh, J.T., Saha, N.R., Rana, D., Sarkar, G., Mollick, M.M.R., Chattoapadhyay, A., Mitra, B.C., Mondal, D., Ghosh, S.K., & Chattopadhyay, D. (2018). Jute cellulose nanofibrils/hydroxypropylmethylcellulose nanocomposite: A novel material with potential for application in packaging and transdermal drug delivery system. *Industrial Crops and Products*, 112, 633-643. <u>https://doi.org/10.1016/j.indcrop.2017.12.069</u>
- [4] Abba, M., Ibrahim, Z., Chong, C.S., Zawawi, N.A., Kadir, M.R.A., Yusof, A.H.M., & Razak, S.I.A. (2019). Transdermal delivery of crocin using bacterial nanocellulose membrane. *Fibers and Polymers*, 20(10), 2025-2031. https://doi.org/10.1007/s12221-019-9076-8
- [5] Peña-Juárez, M.C., Guadarrama-Escobar, O.R., & Escobar-Chávez, J.J. (2022). Transdermal delivery systems for biomolecules. *Journal of Pharmaceutical Innovation*, 17, 319-332. <u>https://doi.org/10.1007/s12247-020-09525-2</u>
- [6] Vega-Vásquez, P., Mosier, N.S., & Irudayaraj, J. (2020). Nanoscale drug delivery systems: from medicine to agriculture. *Frontiers in Bioengineering and Biotechnology*, 8, 79. <u>https://doi.org/10.3389/fbioe.2020.00079</u>
- [7] Wang, Y., Zeng, L., Song, W., & Liu, J. (2022). Influencing factors and drug application of iontophoresis in transdermal drug delivery: an overview of recent progress. *Drug Delivery and Translational Research*, 12, 15-26. https://doi.org/10.1007/s13346-021-00898-6
- [8] Pawar, P.M., Solanki, K.P., & Mandali, V.A. (2018). Recent advancements in

transdermal drug delivery system. International Journal of Pharmacy and Clinical Research, 10(3), 65-73.

- [9] Sarkar, G., Orasugh, J.T., Saha, N.R., Roy, I., Bhattacharyya, A., Chattopadhyay, A.K., Rana, D., & Chattopadhyay, D. (2017). Cellulose nanofibrils/chitosan based transdermal drug delivery vehicle for controlled release of ketorolac tromethamine. *New Journal of Chemistry*, 41, 15312-15319. <u>https://doi.org/10.1039/C7NJ02539D</u>
- [10] Ramadon, D., McCrudden, M.T.C., Courtenay, A.J., & Donnelly, R.F. (2020). Enhancement strategies for transdermal drug delivery systems: current trends and applications. *Drug Delivery and Translational Research*, 12, 758-791. <u>https://doi.org/10.1007/s13346-021-00909-6</u>
- [11] El-Alfy, E.A., El-Bisi, M.K., Taha, G.M., & Ibrahim, H.M. (2020). Preparation of biocompatible chitosan nanoparticles loaded by tetracycline, gentamycin and ciprofloxacin as novel drug delivery system for improvement the antibacterial properties of cellulose based fabrics. *International Journal of Biological Macromolecules*, 161, 1247-1260. <u>https://doi.org/10.1016/j.ijbiomac.2020.06.118</u>

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