JOURNAL OF AGROBIOTECHNOLOGY 2025, VOL 16(2):48-54
e-ISSN: 2180-1983
http://dx.doi.org/10.37231/jab.2025.16.2.407
https://journal.unisza.edu.my/agrobiotechnology/index.php/agrobiotechnology/index



Enhancing the Mechanical Properties of Carrageenan-Tapioca Starch Biocomposite Films for Sustainable Food Packaging

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Received: 02/09/2025, Accepted: 22/10/2025, Available Online: 01/11/2025

ABSTRACT

The global shift toward sustainable materials has spurred research into biodegradable food packaging options. This study explores the improvement of carrageenan-based biocomposite films by using tapioca starch (TS) at various concentrations (0.15 g – 0.30 g). Films were prepared by solution casting, using polyethylene glycol (PEG) as plasticizer and isovanillin as crosslinker. Mechanical, rheological, and structural parameters were investigated, including tensile strength, viscosity, Fourier Transform Infrared (FTIR) spectra and Area Under Peak (AUP) of infrared spectra analysis. The results revealed that carrageenan incorporated with 0.20 g of tapioca starch (TS0.20) produced the highest tensile strength (2.125 MPa) and viscosity (141.55 P), balancing elasticity and film strength. At TS0.20, FTIR analysis with AUP quantification revealed higher intermolecular (20.783) and intramolecular (7.985) OH bonding, as well as better O-SO_3 (13.737) and C=O (10.050) intensities, indicating stronger hydrogen bonding and sulfate-carbonyl interactions. However, higher TS concentrations (more than 0.20 g) resulted in lower AUP values, lower tensile strength, and functional groups intensity values, indicating polymer network breakdown. These findings show that moderate starch incorporation improves mechanical and functional qualities, pointing to carrageenan-tapioca starch films as promising biodegradable food packaging alternatives.

Keywords: Hydrogen bonding, food packaging, carbohydrate polymers, material science, bioresources

INTRODUCTION

The increasing reliance on petroleum-based plastics in food packaging constitutes serious environmental and health risks due to their non-biodegradable nature. Their resilience adds to the growing problem of plastic waste and microplastic contamination, which harms ecosystems and may endanger human health (Khan & Bose, 2024). This worldwide concern highlights the importance of creating sustainable, biodegradable packaging materials as viable alternatives.

Carrageenan, a sulphated polysaccharide derived from red seaweed, has sparked interest due to its film-forming properties, transparency, and biodegradability. However, pure carrageenan films are brittle and very moisture sensitive, making them unsuitable for practical food packaging applications (Wang et al., 2024). Blending carrageenan with other natural polymers is thus regarded as a possible technique for overcoming these constraints (Hamdan et al., 2021). Tapioca starch (TS), which is abundant and inexpensive in Malaysia, has potential as a reinforcing material because of its hydroxyl and carbonyl groups, which can form hydrogen bonds with carrageenan chains. Besides, these combinations of materials have been extensively researched for its film-forming ability, biodegradability, and compatibility with natural additives in biodegradable food packaging.

Previous research indicates that starch incorporation can improve flexibility and tensile strength, but excessive addition may disrupt the carrageenan matrix and decrease barrier function (Praseptiangga et al., 2023). Therefore, research on determining the right starch concentration is crucial. The addition of zein to tapioca starch films increased stiffness and decreased strain at break, improving mechanical strength (Pérez et al., 2021). Tapioca starch improves the biodegradability of films, as seen in blends with polybutylene adipate-co-terephthalate (PBAT), where increasing starch content led to increased breakdown under natural weathering and soil burial conditions (Akhir et al., 2023). These studies show the diverse applications of TS in active and sustainable packaging solutions, highlighting its potential role in Malaysia's agro-food sector.

This article investigates the integration of TS at varied concentrations (0.15-0.30 g) to solve the problem of balancing strength and flexibility in carrageenan-based films. The goals are to evaluate the effects on mechanical, rheological, and barrier properties, as well as to examine molecular interactions, utilizing Fourier-transform infrared (FTIR) spectroscopy with Area Under Peak (AUP) measurement. It is hypothesized that a moderate starch concentration will improve hydrogen bonding and sulfate-carbonyl interactions in carrageenan films, resulting in better tensile strength and viscosity, whereas higher concentrations may disrupt the matrix and increase permeability of biocomposite film.

MATERIALS AND METHODS

Materials

Refined carrageenan was obtained from CV Simpul Agro Globalindo, Indonesia, while tapioca starch (Bidari, Malaysia) was purchased from local supplier. Food-grade crosslinker, isovanillin was obtained from Eternal Pearl, Zhejiang, China and polyethylene glycol (PEG) which act as a plasticizer was bought from Merck, Germany.

Preparation of carrageenan based biocomposite film reinforced by tapioca starch

Refined carrageenan was mechanically stirred with isovanillin in 60 mL distilled water at 60 °C and 1,000 RPM for 50 minutes. Next, tapioca starch powder (0 to 30 g) and PEG solution were added and mixed for a total of 60 minutes in order to obtain a homogeneous solution. The samples were labelled as Control, TS0.15, TS0.20, TS0.25, and TS0.30 where the number represents the weight (gram) of tapioca starch. All biocomposite film formulations were shown in Table 1. Upon drying, the viscosity of formulation solution was analysed. Then, the formulation solution was casted on a stainless-steel tray and left dry at room temperature for three (3) days. The dried film was then employed for characterization and analysis.

Table 1. The formulation with raw materials of refined carrageenan, tapioca starch, polyethylene glycol (PEG), isovanillin, and distilled water.

Formulation sample	Refined carrageenan (g)	Tapioca starch (g)	PEG (mL)	Isovanillin (g)	Distilled water (mL)
Control	1	0	0.5	0.02	60
TS0.15	1	0.15	0.5	0.02	60
TS0.20	1	0.20	0.5	0.02	60
TS0.25	1	0.25	0.5	0.02	60
TS0.30	1	0.30	0.5	0.02	60

*In formulation sample, the number represents the weight (gram) of tapioca starch.

Analysis of formulation solution and biocomposite film reinforced by tapioca starch

Viscosity analysis of formulation solution

The viscosity of the formulation solutions was analysed using a ViscoQC 100L viscometer (Anton Paar, GmbH, USA) equipped with L4 probe. The usage of the probe was based on the solution of the films, where the viscosity was not more than 200 P. Approximately 15 mL of the sample solution was programmed at speed of 100 rpm and at a constant temperature of 60^{o}C in triplicate.

Tensile strength analysis of biocomposite film

The tensile strength of 1.5 cm x 10 cm strips of bicomposite films were determined using an electronic TA.XTplusC Texture Analyser (Stable Micro Systems, United Kingdom) fitted with a 5 kg load cell. The method was in accordance with ASTM D882-12 and a crosshead speed of 50 mm/s was employed. The tensile machine was operated by using Spaghetti/Noodle probe with some modifications to fit the size of film sample.

Functional group analysis of biocomposite film

Fourier transform infrared spectroscopy (FTIR) was conducted to examine chemical bonding occurred in the carrageenan biocomposite film. An IRPrestige-21 spectrometer (Shimadzu Corporation, Japan) was used with the spectra range of 650 to 4000 cm^{-1}. A total of 32 scans were acquired and with a spectral resolution of 8 cm^{-1}. The spectra were analysed using IRsolution software.

Statistical analysis

Each experiment was repeated in triplicate to ensure a statistically valid result were produced. The data were analyse using Microsoft Excel Professional Plus 2021 and OriginPro 9.0 to determine the average, significant difference, and graph plotting for each data. To compare the means, One-Way Analysis of Variance (ANOVA): Post Hoc Multiple Comparison, Duncan Test with confident interval of 95% was conducted using Microsoft Excel Professional Plus 2021.

RESULTS AND DISCUSSION

Viscosity of formulation solution and tensile strength of carrageenan biocomposite film reinforced by tapioca starch

The connection between viscosity and tensile strength in carrageenan film biocomposite with different tapioca starch (TS) concentrations is shown in Fig. 1. As 0.15 g of TS is added, the viscosity of the control formulation

drops considerably to about 122.18 P from its initial peak of 155.48 P. Viscosity gradually decreases as TS concentration increases from 0.20 g to 0.30 g, reaching the control value at TS0.30 with 91.15 P. On the other hand, the addition of TS0.15 decreases the tensile strength of the control formulation in comparison to Control formulation, from 2.919 MPa, to 1.545 MPa. The tensile strength progressively decreases as TS concentration rises to 0.30 g, stabilizing at 1.347 MPa and 1.309 MPa. It is noteworthy to consider that the film with TS0.20 has the maximum tensile strength at 2.125 MPa.

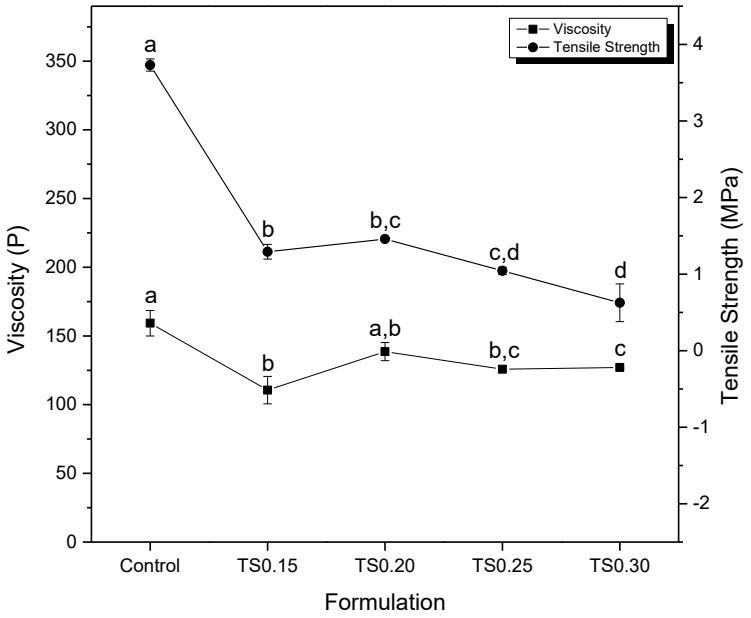


Fig. 1. Viscosity of formulation solution and tensile strength of carrageenan biocomposite film reinforced by tapioca starch.
[a,b,c] Denote significant differences between formulations.

The viscosity of the carrageenan solution constantly decreases when tapioca starch is added. This reduction in viscosity is likely due to starch disrupting the carrageenan network, weakening molecular interactions and lowering flow resistance (You et al., 2021). Higher TS concentrations, such as TS0.25 and TS0.30, are where this disruption is most obvious as the viscosity significantly decreases at these concentrations because of less cohesive contacts between polymer chain and the starch properties enhance the liquid-like properties of the gel and biocomposite (Sheng et al., 2018a). When TS is added, the tensile strength of carrageenan films also decreases. This study suggested that the hydroxyl-rich tapioca starch (TS) competes for or redistributes hydrogen bonding sites within the carrageenan network, forming new hydrogen bonds with both carrageenan and water molecules (as supported by the FTIR analysis in the following section) (Prasetyaningrum et al., 2024; Sheng et al., 2018b). This competition weakens the original carrageenan–carrageenan hydrogen bonding network, thereby reducing viscosity and tensile strength at higher starch concentrations (Notonegoro et al., 2024). However, at TS0.20, the author proposed that the tensile strength slightly increases, possibly as a result of the starch and carrageenan forming an ideal and stronger hydrogen bonding interaction. This implies a balance where starch serves as a plasticizer to improve elasticity and elongation at break, hence enhancing mechanical characteristics. Lowest tensile strength at TS0.30 indicates a structural failure caused by the incompatibility of the two materials. In general, maximizing the mechanical characteristics of starch-based biocomposite in food packaging applications requires a knowledge of these relationships.

Functional group determination of carrageenan biocomposite film reinforced by tapioca starch

The vibrational modes of functional groups are affected differently by different tapioca starch (TS) concentrations, as demonstrated by the FTIR spectrum analysis of biocomposite films in Fig. 2. Important peaks for hydroxyl groups (O-H stretching) are located about 3,378 cm^{-1}; these peaks become more intense at

increased TS concentrations, indicating an increase in hydroxyl groups. Peaks at 2,931 cm^{-1} for hydroxymethyl groups (CH_2-OH) and 1,240 cm^{-1} for sulphate groups (O-SO_3) exhibit differences in intensity that are suggestive of structural modifications in the polymeric matrix carried by the addition of starch. Notable peaks can be found at 844 cm^{-1} for sulphate ester groups (C-O-SO_4) and 922 cm^{-1} for carbonyl groups (C=O), where intensity changes imply that starch inclusion alters the structure of the carrageenan network.

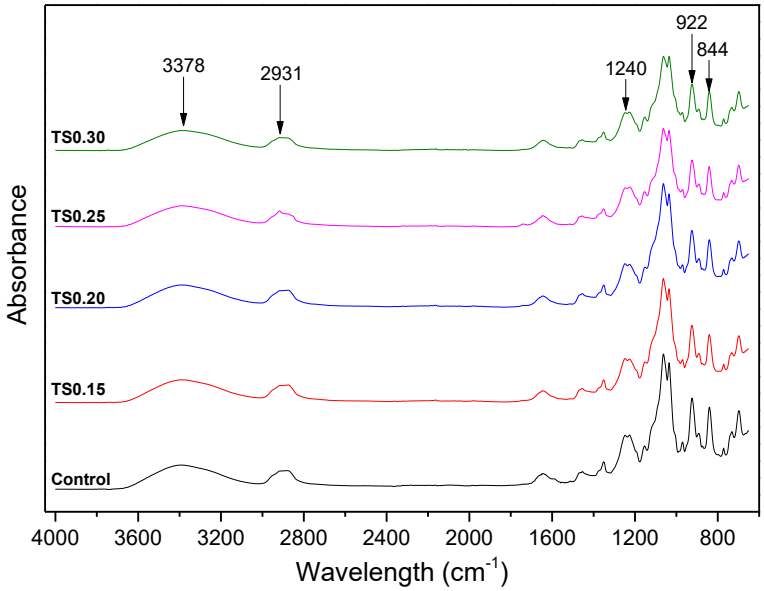


Fig. 2. Infrared spectroscopy of carrageenan biocomposite film reinforced by tapioca starch.

Table 2 presents a quantitative analysis of these functional categories using the Area Under Peak (AUP). The peak area was determined by integrating the peak area at specific wavelength range (Hamdan, 2022; Mohd Amin, 2016). Intermolecular hydrogen bond refers to a hydrogen bond (H-bond) occurs between separate molecules in a substance, while intramolecular hydrogen bond refers to that H-bond that hold atoms together within a single molecule (Domingo & Pérez, 2025).

Table 2. Peak area of major functional groups in carrageenan biocomposite film reinforced by tapioca starch.

Functional Group	**Intermolecular OH**	**Intramolecular OH**	**(O-SO_3)**	**(C-O-SO_4)**	**(C=O)**
Wavelength range	3500-3200	3200-2700	1315-1175	865-810	960-900
Control	14.502	4.201	13.593	8.111	10.122
TS0.15	19.384	7.145	13.176	7.809	9.81
TS0.20	20.783	7.985	13.737	8.029	10.05
TS0.25	18.479	6.038	11.857	6.847	8.487
TS0.30	17.929	5.745	11.492	6.723	8.41

The hydroxyl group peak first increases with the addition of tapioca starch. Yet, it then falls at higher TS concentrations, possibly as a result of complex formation or saturation limiting free hydroxyl groups, according to the AUP data. Hydrogen bonds form between tapioca starch's hydroxyl groups and other components including carrageenan, chitosan, and plasticizers, which improves compatibility and interaction within the composite matrix (Noviagel et al., 2024). This bonding is critical for improving the tensile strength and Young's modulus of the biocomposites, as indicated by higher mechanical characteristics when additives and toughening agents are incorporated. This work suggested that the occurrence of higher tensile strength of biocomposite film

reinforced by tapioca starch in TS0.20 (as shown in Fig. 1) was supported by the result of stronger intermolecular and intramolecular hydrogen bonding interaction as presented in Table 2 (Adam et al., 2021).

Corresponding patterns are seen for the sulphate and carbonyl ester groups, where declines occur after initial increases in concentration, suggesting interactions that reduce the availability of free functional groups. Nonetheless, the hydroxymethyl group exhibits a significant initial increase in AUP upon TS addition, which is then followed by a decline, indicating that increased starch concentrations can cause complex formation that lowers free hydroxymethyl groups and reduce the effectiveness of hydrogen bonding in thermoplastic starch together with its mechanical properties (Nurhajati et al., 2019). These results demonstrate the complex interactions between starch and carrageenan, which affect the distribution of functional groups in the biocomposite film and, in addition, its mechanical and structural characteristics.

CONCLUSION

The increasing prevalence of petroleum-based plastics creates significant environmental challenges, emphasizing the need for biodegradable alternatives in food packaging. To address these concerns, this work developed carrageenan-tapioca starch (TS) biocomposite films and examined how different starch concentrations affected film performance. TS0.20 outperformed the other formulations in terms of tensile strength (2.125 MPa) and viscosity (141.55 P). FTIR-area under peak analysis revealed increased intermolecular and intramolecular hydrogen bonding, as well as greater sulfate-carbonyl interactions, at this concentration. In contrast, TS0.30 had the poorest performance due to network disruption and fewer functional group contacts, demonstrating the adverse consequences of excessive starch inclusion. These findings highlight the importance of modifying starch concentration to achieve optimal mechanical strength, barrier characteristics, and molecular stability. The study validates carrageenan-TS films' potential as eco-friendly packaging materials, which is especially significant in Malaysia, where both raw materials are easily available. Future research should look into nanofiller reinforcement, long-term biodegradability in different environments, and real-food packaging studies to validate industrial applicability. Optimized carrageenan-TS films could serve as viable alternatives to plastics, supporting global sustainable packaging initiatives.

ACKNOWLEDGMENTS

This research was funded by the Ministry of Higher Education (MOHE) Malaysia under the Fundamental Research Grant Scheme FRGS/1/2024/STG05/UNISZA/02/1; RF036.

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How to cite this paper:
Bakri, M. A. M., Adnan, K. S. M., Hamdan, M. A., (2025). Enhancing the mechanical properties of carrageenan-tapioca starch biocomposite films for sustainable food packaging. Journal of Agrobiotechnology, *16*(2), 48-54