REKAYASA 23 (2) (2025): 94-104

REKAYASA

Jurnal Penerapan Teknologi dan Pembelajaran

http://journal.unnes.ac.id/journals/rekayasa

Effect of Ultrasonic Processing on Mechanical and Surface Properties of Chitosan-Alginate-Starch Edible Films

Desi Rahmawati*[1], Azafilmi Hakiim[1], Indah Cintami[1], Alehandro Sitinjak[1]

[1]Department of Chemical Engineering, Universitas Singaperbangsa Karawang, Karawang, Indonesia

*Email: 2210631230023@student.unsika.ac.id

DOI: https://doi.org/10.15294/rekayasa.v23i2.49226

Abstract

Marine plastic waste has driven the development of biodegradable packaging alternatives such as edible films. However, their mechanical strength and structural homogeneity remain challenging. This study investigates the effect of ultrasonic processing on the mechanical and surface properties of edible films based on chitosan–alginate–starch blends. Edible films were prepared with various polysaccharide combinations and subjected to ultrasonic treatment during processing. The sample was treated with ultrasonic waves at a frequency of 37 kHz and a temperature of 30 °C for 3 minutes. The results show that ultrasonic treatment improved surface homogeneity, as indicated by smoother structures and reduced bubble formation compared to non-ultrasonic films. In terms of mechanical properties, ultrasonic treatment exhibited varying effects on tensile strength but consistently increased elongation at break, indicating enhanced flexibility. The highest tensile strength was observed in chitosan-based films (45.479 MPa), while the highest elongation (41%) was achieved in chitosan–alginate films under ultrasonic treatment. The ternary blend (chitosan–alginate–starch) demonstrated balanced mechanical properties in both ultrasonic and non-ultrasonic conditions. Chemical analysis using FTIR spectroscopy showed that ultrasonic treatment affected intermolecular interactions within the film matrix, as indicated by shifts and changes in the intensity of absorption bands in several functional groups. These findings suggest that ultrasonic processing plays a crucial role in improving film structure and flexibility, while the polysaccharide composition governs the balance between strength and elasticity.

Keywords: Biodegradable Packaging, Elasticity, Film Structure, Marine Waste, Polysaccharide Combinations.

INTRODUCTION

Marine plastic debris is an environmental issue that has attracted attention from many countries, including Indonesia. According to a November 2021 report by the International Union for Conservation of Nature (IUCN), more than 300 million tons of plastic are produced annually for various purposes. Approximately 14 million tons of this plastic enters the oceans. Plastic waste accounts for approximately 80% of all marine debris, both on the surface and in the deep sea (Kurtela & Antolović, 2019). Based on this data, marine waste is dominated by plastic, making it a serious threat to the marine ecosystem (Rezasyah & Sari., 2022). Indonesia ranks second as a contributor of plastic waste to the ocean after China. The volume of plastic waste reaches 64 million tons per year, of which 3.2 million tons are estimated to end up in the ocean (Kosior & Crescenzi 2020). A total of 109 rivers are the main routes for plastic waste to enter the ocean and 53 of them contribute around 80% of global plastic emissions (Meijer et al., 2021).

The Indonesian food industry still relies on plastic packaging, with usage reaching approximately 80% (Pramiati, 2021). The plastic packaging market reached USD 348.9 billion in 2023 and is projected to continue growing at approximately 4.2% through 2030 (Başboğa, 2023). The year-over-year increase has the potential to cause greater environmental impacts if not addressed early. Edible film is one of the innovations being developed, edible packaging made from a mixture of various agricultural products rich in carbohydrates (Wu et al., 2024).

Edible film is known as a packaging material that is safe to use, environmentally friendly, and easily biodegradable. Edible film protects food from microorganisms, such as bacteria and mold, making it safer, less prone to spoilage, and extending the product's shelf life (Li et al., 2024). This product supports efforts to provide healthy, safe, high-quality, reliable, and affordable food for the society (Biesbroek et al., 2023). Edible films act as a barrier that can help preserve products by inhibiting the transfer of substances such as moisture, oxygen, fats, and dissolved substances, thereby extending the product's shelf life (Kumar et al., 2022).

Regarding the synthesis and characterization of edible films based on cassava starch and chitosan with the addition of honey as a natural plasticizer, the films exhibited tensile strengths ranging from 0.987 to 2.325 MPa and elongation at break ranging from 25-72% (Sapria et al., 2025). Improvements of thermal and mechanical properties of achira starch/chitosan/clay nanocomposite films with ultrasonic processing yielded a tensile strength of 1.72 – 4.21 Mpa and an elongation at break of 10.42%-63.66% (Aguirre-Laredo et al., 2023) This combination generally improves the film's mechanical properties, but still has limitations in achieving an optimal balance.

Ultrasound generates high pressure and strong shear forces that can break the bonds between nanoparticles, thereby reducing agglomeration. This process serves to prevent the formation of clumps that can reduce the barrier properties. (Lan et al., 2020). Ultrasonic process will make the polymer structure break down more easily through natural processes (Kirsh et al., 2020). In chemical processes, ultrasonic energy does not interact directly with the sample but rather through an intermediate medium, such as water. This technology has been proven to reduce processing time and minimize the need for solvents, thereby making the process more efficient (Sun et al., 2025). Sound waves generated from electrical energy via a transducer then propagate through the liquid medium and are transmitted to the sample via a cavitation mechanism (Rezk, 2021). The cavitation effects caused by ultrasound are divided into steady-state cavitation and transient cavitation (Zhang et al., 2023). Cavitation is a process in which small bubbles form in a medium; these bubbles eventually grow larger and then collapse, releasing a large amount of energy (Sarraf et al., 2022).

Ultrasonic processing of edible films not only produces a smoother surface but can also improve the material's microstructure. Through the phenomenon of cavitation, ultrasound improves component dispersion by breaking down aggregates, resulting in a more even particle distribution. This reduces void formation in the matrix and results in a more homogeneous film structure. A more uniform structure allows for more efficient stress transfer when the film is subjected to tensile forces. As a result, elongation tends to increase due to better flexibility, while tensile strength can vary depending on the process conditions and the resulting structure (Guruchandran et al., 2024). This increase in homogeneity generally results in increased elongation due to improved material homogeneity, along with a more even stress distribution during solidification. However, the effect on tensile strength is not consistently increased, as ultrasonic intensity and changes in internal structure can cause variations in material strength, including possible degradation or changes in crystallinity. The use of multicomponent blend systems such as alginate–chitosan–starch is a strategy for obtaining balanced mechanical properties, because the combination of these biopolymers can improve material properties such as size, shape, orientation, distribution, and concentration of reinforcement in the matrix, and produce an optimal combination of tensile and elongation (Aguirre-loredo et al., 2023).

Chitosan is a derivative of chitin with a reduced degree of acetylation and is classified as an copolymer α-(1→4) glucosamine $(C_6H_{11}O_4N)n$ with varying numbers of N-acetyl groups. This compound is a white to pale pink solid that is insoluble in water but soluble in weak acids (Al-rooqi et al., 2022). Chitosan has broad-spectrum antimicrobial activity and is effective in inhibiting the growth of various pathogenic microorganisms, including gram-positive bacteria, Gram-negative bacteria, and fungi (Zhanifathul et al., 2025).

Alginate is one of the primary polysaccharides that make up the cell walls of brown algae. Alginate consists of linear copolymer chains composed of units of guluronic acid (G) and manuronic acid (M); this composition influences the physical properties of alginate, such as gel strength, viscosity, and mechanical stability (Abka-Khajouei et al., 2022).

Potato starch (*Solanum tuberosum L.*) is an ideal material for making edible films because it is transparent, odorless, and tasteless, making it similar to the synthetic polymers commonly used in food packaging (Soto et al., 2025). Starch consists of two main polymers that cannot be separated by hot water: amylose, which has a linear α-1,4 structure, and amylopectin, which has a branched structure with bonds α-1,6 (Guo et al., 2023).

This study presents a more comprehensive approach compared to previous studies, which generally utilized only a combination of two types of polysaccharides. In this study, chitosan, alginate, and potato starch were examined in various formulations, including the use of each material individually, two-component combinations, and three-component combinations. In addition, the effect of the ultrasonic process on each formulation was evaluated to determine its role in improving the characteristics of the resulting edible films. This approach is expected to provide a deeper understanding of the role of intermolecular interactions in film matrix formation and their relationship to the mechanical properties and surface characteristics of edible films. Based on this, study aims to develop edible films based on chitosan-alginate-starch and to evaluate the effects of variations in ingredient composition and the ultrasonic process on the quality of the resulting films. The combination of these three polysaccharides with the ultrasonic process is expected to produce effects that enhance mechanical properties, improve structural homogeneity, and result in a more uniform film surface. The results of this study are expected to contribute to scientific understanding of the use of polysaccharide-based hydrocolloids as components of biodegradable food packaging. In addition, this study is also expected to provide a scientific basis for the use of ultrasonic technology in improving the characteristics of edible films, thereby supporting the development of more environmentally friendly packaging materials with the potential for industrial-scale application.

METHOD

The materials used were chitosan, alginate, potato starch, 1% acetic acid, and distilled water. The equipment used in this study included beakers, glass funnels, volumetric pipettes, droppers, measuring cylinders, stirring rods, petri dishes, an analytical balance, an ultrasonic device, a hotplate, a magnetic stirrer, and a fan.

The edible films were prepared using seven different formulations, consisting of single-component, two-component, and three-component formulations. The composition ratio of chitosan, alginate, and potato starch was determined based on polymer mass. Each polymer was used in an equal mass of 1 g, resulting in a chitosan:alginate:potato starch ratio of 1:1:1 (w/w/w). The polymers were dissolved separately using different volumes of solvent; chitosan in 25 mL of acetic acid, while alginate and potato starch were each dissolved separately in 10 mL of distilled water. The potato starch solution was then heated to 120°C to achieve gelatinization. After all ingredients were dissolved and formed a homogeneous solution, each solution was mixed according to the predetermined formulation. Each formulation was then divided into two groups: a group treated with ultrasonication and a group without ultrasonication. Ultrasonication was performed at a frequency of 37 kHz and a temperature of 30°C for 3 minutes. Next, the film solutions were poured into plastic petri dishes and dried at room temperature using airflow from a fan until all the solvent had evaporated and a dry edible film layer had formed.

The characterization of the edible film was conducted through mechanical property testing and functional group analysis. Mechanical property testing, which included tensile strength and elongation at break, using a Brookfield CT3 with specimens measuring 30 mm x 5 mm. Meanwhile, functional group analysis was performed using Fourier Transform Infrared Spectroscopy (FTIR) to identify intermolecular interactions occurring in the edible film matrix due to variations in formulation and the ultrasonic process.

RESULTS AND DISCUSSION

Surface Analysis

The surface analysis of the edible film in this study was conducted based on observations of its physical appearance after the drying process. Surface characteristics serve as an important indicator for evaluating the homogeneity and quality of the resulting film. The results of the edible film are shown in Figure 1. The figure shows that the edible film without ultrasonic treatment still contains many trapped air bubbles, resulting in a less even surface. In contrast, the edible film treated with ultrasonic treatment has a smoother and more homogeneous surface with significantly fewer bubbles. This difference indicates that the use of ultrasonication plays a role in increasing dispersion, resulting in a more uniform film structure. Ultrasonic technology improves food quality through acoustic cavitation, which involves the rapid formation and collapse of bubbles, generating local shear forces and high-speed jets (microjets) that impact particles, resulting in changes in the food matrix (Prempeh et al., 2025).

Tensile Strength

Tensile strength testing is designed to evaluate the ability of edible films to withstand applied loads or tensile forces (Shah et al., 2023). Edible films with high tensile strength demonstrate superior resistance to pressure and mechanical stress, thereby providing effective protection for food products against physical damage during handling, storage, and distribution (Feng et al., 2025).

Furthermore, ultrasonic treatment can improve the dispersion and homogeneity of a solution through the phenomenon of cavitation, thereby strengthening intermolecular interactions in some formulations; however, in certain formulations, it can also cause partial degradation of the polymer chains, resulting in varying tensile strength values (Kliem et al., 2020).

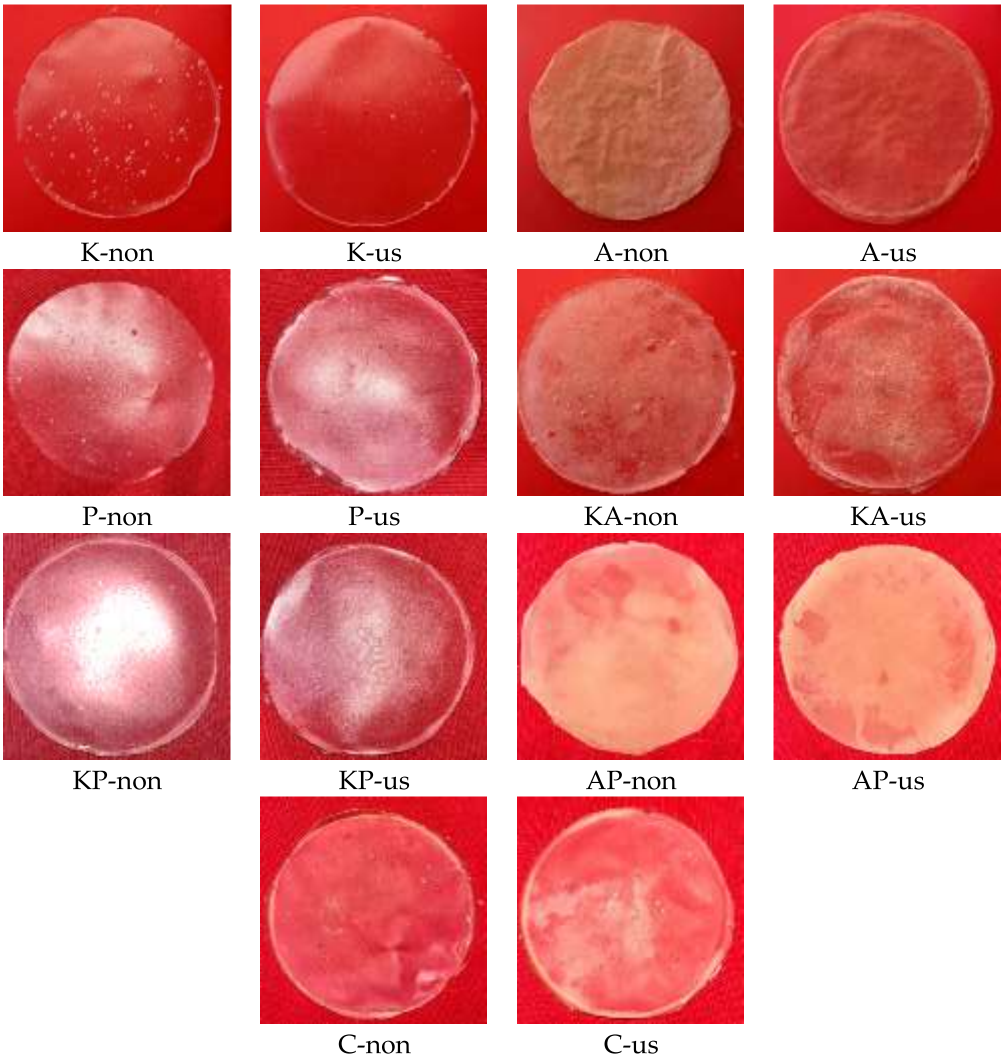


Figure 1. Edible Film Samples with Various Combinations

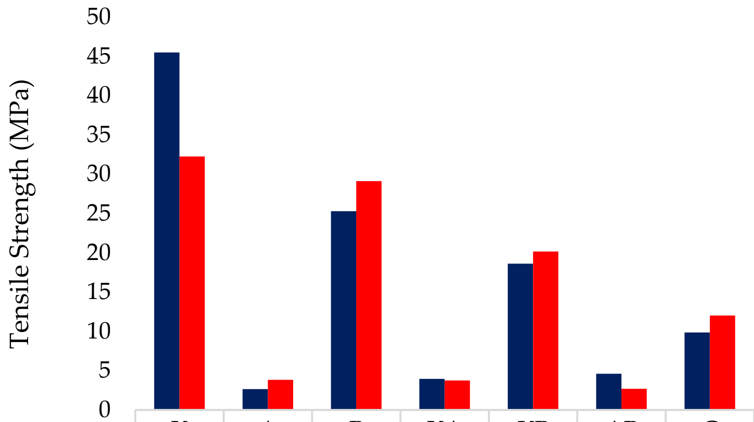
The variation in tensile strength values among samples is the result of a combination of the intrinsic properties of each polymer. The chitosan sample with ultrasonic processing has the highest tensile strength value among the others, at 45.479 MPa. This value is higher than the result reported by Sutharsan et al., 2023 who obtained a tensile strength of approximately 27 MPa for chitosan films dried at 21°C. This comparison indicates that the tensile strength of the chitosan film in this study increased by approximately 68% compared to the value reported in the previous study. These results suggest that the preparation conditions used, including ultrasonic treatment, have the potential to improve structural compactness and strengthen intermolecular interactions within the film matrix. The ultrasonic process enhances the homogeneity of the film-forming solution, reduces particle aggregation, and strengthens interactions between chitosan chains through the formation of a denser and more uniform network.

Research conducted by Badwan et al. (2015) states that an increase in the percentage of chitosan in the formulation is accompanied by an increase in the force required to break the specimen. This occurs because stronger hydrogen bonds form in pure chitosan samples, making the interchain bonds more difficult to break. The formation of strong and stable bonds between the constituent molecules of the edible film directly contributes to the high tensile strength of the film (Tafa et al., 2023). A higher chitosan content in the film results in greater structural strength compared to a structure composed solely of alginate (Wibowo et al., 2023).

Elongation at Break

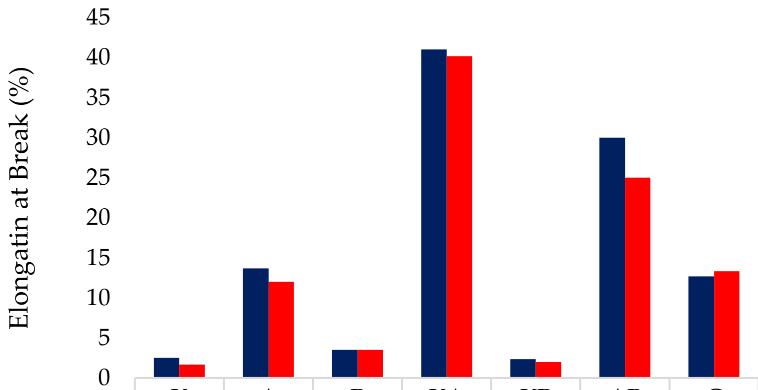
Elongation at break indicates the extent to which a film can stretch before reaching its breaking point; therefore, this parameter is related to the film's flexibility and elasticity (Yuan et al., 2022). Figure 3 shows that ultrasonic treatment tends to increase the elongation value, making the film more flexible and capable of stretching further before breaking. The sample with the highest elongation value was the chitosan-alginate formulation treated with ultrasonication, at 41%. The improvement in the elastic properties of chitosan-alginate films is due to the formation of molecular interactions via a polyelectrolyte complex mechanism between the $NH3+$ groups on chitosan and the COO- groups on alginate (Hamedi et al., 2019).

Based on the evaluation of mechanical properties using tensile strength and elongation as parameters, the composite samples exhibited relatively balanced values for both the ultrasonic-treated (9.867 MPa and 12.667%) and non-ultrasonic-treated (12.021 MPa and 13.33%) samples. This condition differs from other samples, which tend to exhibit a trade-off, where high tensile strength values result in low elongation values (Kholiq & Kusuma, 2024). The percentage of elongation shows an inverse relationship with tensile strength. An increase in the percentage of elongation is generally accompanied by a decrease in tensile strength, indicating that the material is becoming more elastic (Tafa et al., 2023). For example, the chitosan sample achieved high tensile strength (45.479 MPa) and low elongation (2.5%), resulting in stiffness and low elasticity. Furthermore, the chitosan-alginate sample achieved low tensile strength (3.948 MPa) and high elongation (41%), indicating elastic properties but low strength. The balance of tensile strength and elongation values in the blended samples indicates that the resulting film has quite good mechanical properties: it is neither too stiff nor easily brittle.



	K	A	P	KA	KP	AP	C
US	45,479	2,646	25,286	3,948	18,597	4,596	9,867
Non-US	32,264	3,825	29,122	3,737	20,170	2,690	12,021

Figure 2. Results of The Tensile Strength Analysis of Edible Film



	K	A	P	KA	KP	AP	C
US	2,500	13,667	3,500	41,000	2,333	30,000	12,667
Non-US	1,667	12,000	3,500	40,167	2,000	25,000	13,333

Figure 3. Results of The Elongation at Break Analysis of Edible Film

Fourier Transform Infra-Red (FTIR) Analysis

FTIR is an analytical technique used to identify molecular structures simultaneously and quantitatively. This technique produces characteristic spectral patterns and absorption peaks that can be used to identify molecular structures and chemical bond types. FTIR data interpretation requires the support of a reference database for comparison (Nandiyanto et al., 2023).

The molecular interaction of the blend film (chitosan-alginate-potato starch) with ultrasonic and non-ultrasonic treatments is shown in Figure 4. The O-H group appears at 3536 cm-1 absorption in the non-ultrasonic treatment and 3531 cm-1 in the ultrasonic treatment. The O-H group formed indicates the presence of the constituent components of each material. The O-H group in chitosan comes from chitin after degassing to produce O-H and N-H groups (Muchtaromah et al., 2023), in alginate, the O-H groups are derived from manuronic acid and guluronic acid (Sari et al., 2023), meanwhile in starch, the strong absorption intensity of the O-H groups is due to the large number of O-H groups involved in linking the amylose and amylopectin structures (Kumar et al., 2022).

Chitosan exhibited a characteristic N–H stretching vibration at 3484 cm^{-1} in the non-ultrasonic treatment. Following ultrasonic treatment, this absorption band shifted to a lower wavenumber (3433 cm^{-1}), indicating enhanced intermolecular interactions. Alginate showed characteristic carboxylate (COO^-) absorption bands corresponding to the symmetric stretching vibration at 1516 cm^{-1} and the asymmetric stretching vibration at 1656 cm^{-1}. After ultrasonic treatment, these bands shifted to 1511 cm^{-1} and 1635 cm^{-1}, respectively, suggesting changes in the ionic environment of chitosan and alginate and indicating stronger ionic interactions between the two biopolymers (Hamedi et al., 2019).

The C–H stretching vibration was observed at 2423 cm^{-1} for the non-ultrasonic treatment and shifted slightly to 2410 cm^{-1} after ultrasonic treatment, confirming the presence of glucose-derived structural components. The presence of starch was further confirmed by the characteristic C–O stretching vibration, which appeared at 1135 cm^{-1} in the non-ultrasonic sample and shifted to 1158 cm^{-1} following ultrasonic treatment. This C–O ether linkage is characteristic of the amylose fraction within the starch structure (Šárka et al., 2023).

The observed shifts in wavenumber and changes in the intensity of characteristic absorption bands indicate stronger hydrogen bonding among the edible film components, resulting in a more stable and homogeneous polymer network (Li et al., 2022). The shift toward lower wavenumbers after ultrasonic treatment is attributed to acoustic cavitation, where the collapse of microscopic bubbles generates localized shear forces and microstreaming that disrupt weak intermolecular interactions and promote a more uniform dispersion of polymer chains. Consequently, stronger intermolecular interactions are established, producing a denser and more homogeneous edible film matrix (Aguirre-Loredo et al., 2023). Moreover, the absence of new absorption bands indicates that ultrasound did not generate new functional groups but instead reorganized the polymer network by strengthening the existing intermolecular interactions within the edible film matrix.

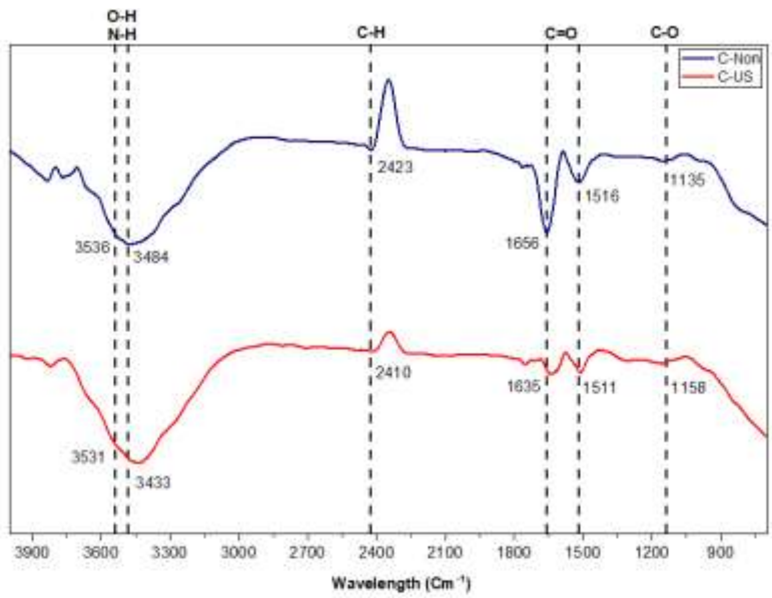


Figure 4. FTIR Spectrum of a Blended Edible Film Formulation

CONCLUSION

The formulation and combination of biopolymers significantly influenced the properties of the edible films. Single-component formulations and binary blends exhibited a trade-off between tensile strength and elongation at break, whereas the ternary blend provided a more balanced combination of mechanical properties. Ultrasonic processing enhanced the homogeneity of the film matrix by promoting stronger intermolecular interactions and a more uniform polymer network, resulting in a smoother surface and improved structural integrity. The chitosan–alginate–starch ternary blend was identified as the optimum formulation, providing the best balance between tensile strength and elongation at break. Future studies should investigate different component ratios, evaluate food packaging applications, and optimize ultrasonic processing conditions to further improve film performance.

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