

# Synthesis and Characterization of Green Thermoplastic Composite from Renewable Resources

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## ABSTRACT

Due to concerns regarding poor degradability and the potential for environmental contamination, the utilization of petrochemical-based plastics should be actively discouraged, leading to a shift toward the adoption of biobased plastics derived from natural sources. Biobased plastics, sourced from renewable materials, are characterized by their elevated costs, biodegradability, compostability, non-toxicity, and minimal environmental impact. This study investigates the potential of utilizing potato waste starch in the production of bioplastics specifically for packaging applications. In this research, the polymer matrix derived from potato waste was modified with chitosan to enhance its antimicrobial, antifungal, and antiviral properties. Such modifications are expected to significantly improve the biocompatibility, bioadhesiveness, and non-toxicity of the potato waste polymer. Consequently, a modified polymer matrix known as chitosan-modified polylactic acid plastics was synthesized. To further augment the mechanical properties of these bioplastics, a jute fiber reinforcement was incorporated into the polymer matrix. The characterization of the bioplastics was conducted using Fourier-transformed infrared spectroscopy (FTIR), while the tensile properties of the plastic composites were evaluated through tensile testing procedures. The findings indicated that the tensile strength of the composites exhibited an increase in correlation with the concentration of the modifier. Conversely, a significant reduction in elongation at break was observed as the concentration of the modifier increased, highlighting the trade-off between strength and flexibility in the modified bioplastics.

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## INTRODUCTION

Plastic is becoming an integral part of life because of its wide range of applications. Petroleum-based plastics, such as polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), and polystyrene (PS), have been widely used in packaging and other day-to-day life (Akbar *et al.*, 2019). The large production and utilization of synthetic polymers have accumulated plastic waste, creating a serious environmental threat. These environmental threats necessitate eco-friendly plastics of natural origin that are biodegradable, non-toxic, and cost-effective (Bashir *et al.*, 2021). Biobased plastics are produced from biomass biopolymers such as starch, cellulose (Figure 1), and proteins (Akbar *et al.*, 2019). Sweet potato is one of the sources of starch-based plastics, converted to glucose by fermentation reaction and further converted to polylactic acid, an important polymer to produce bio-based plastics.

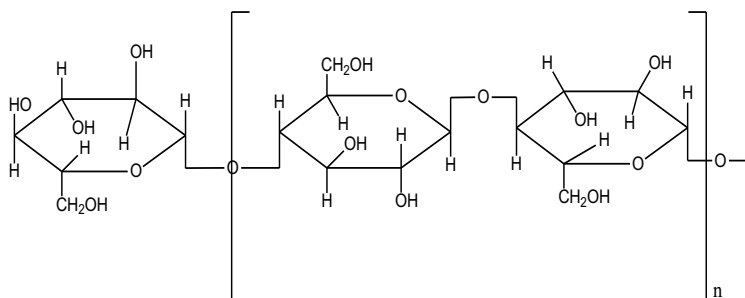


Fig 1 Chemical Structure of cellulose

Polylactic acid is a thermoplastic that is so promising in a wide range of industrial and packaging applications (Bashir *et al.*, 2021). However, starch-based plastics exhibit certain deficiencies, such as brittleness, poor mechanical strength, and sensitivity to moisture. These can be improved by modification using biopolymer. Chitosan is the most qualified candidate due to its unique properties, such as biodegradability, non-toxicity, anti-microbial, anti-fungal, and biocompatibility. Hence, it can be used to improve the thermal stability, plasticity, and antimicrobial properties of biobased plastics (Bashir *et al.*, 2021)

However, starch-based bioplastic has some drawbacks, such as low physical properties, brittleness, and water resistance due to starch's hydrophilic properties. Chitosan is added to the polymer matrix to improve the properties of biobased plastic (Tuly *et al.*, 2017). Chitosan can be added to starch-based bioplastic due to its hydrophobic nature compared to starch. Chitosan is one of the most attractive sustainable biopolymers due to its availability and remarkable intrinsic properties, including digestibility, bacteriostatic and anti-inflammatory effects, biocompatibility, and biodegradability (Nguyen *et al.*, 2021).

Chitosan is a natural cationic amino polysaccharide with glucosamine units linked by 1, 4- $\beta$ -glycosidic bonds (Jayakumar *et al.*, 2011). It is obtained by alkaline deacetylation of chitin, the main component of the exoskeleton of crustaceans (Mohd Zaid *et al.*, 2015). Chitosan is one of the most attractive sustainable biopolymers due to its availability and remarkable intrinsic properties, including digestibility, bacteriostatic and anti-inflammatory effects, biocompatibility, and biodegradability (Gumel *et al.*, 2018). Chitosan is a natural cationic amino polysaccharide with glucosamine units linked by 1, 4- $\beta$ -glycosidic bonds. It is obtained by alkaline deacetylation of chitin, the main component of the exoskeleton of crustaceans (Mohd Zaid *et al.*, 2015). Polylactic acid can be modified via an amidation reaction between the amino group of the main chain of chitosan and the terminal carboxyl group of the polylactic acid (Layla *et al.*, 2018).

The copolymer blends of chitosan polylactic have shown some drawbacks when used as a substitute for plastics in packaging applications. These deficiencies include poor tensile strength, high elongation to break, and thermal instability. To improve such conditions, natural fibers obtained from Cellular origin, like jute, kenaf, hemp, and baobab, can be used to reinforce the copolymer matrix. This study aims to prepare bio-based plastic composite film reinforced with Jute fiber for packaging applications.

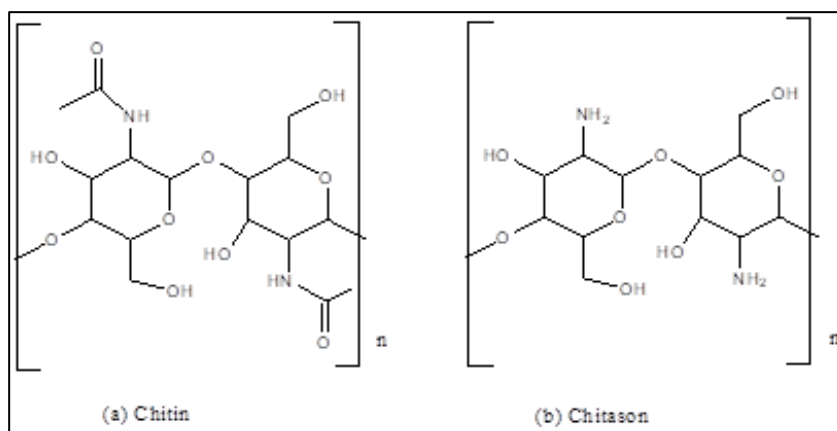


Fig. 2: Structure of (a) Chitin, (b) Chitosan

## EXPERIMENTAL

### Reagents and Materials

The potato waste used for starch production was obtained from a local potato cheap vendor at Kabuga Dala Local Government, Kano State. The Snells shells used to synthesize chitosan were obtained from the Wudil Local Government Sea site, Kano State. In contrast, the Jute fiber was obtained from the Dan Zaki Village, Gezawa Local Government, Kano State plant. This research uses HCl, NaOH, and Acetic acid (Sigma Aldrich), which were procured from the Polymer Chemistry Laboratory, Bayero University Kano. Potato dextrose Agar was obtained from the Microbiology Laboratory at Bayero University Kano. All other chemicals used were grade analytical reagents: ethanol, glycerol, sorbitol, THF, and ammonium oxalate.

### Synthesis of chitosan from sea waste (Snell shells)

Chitosan was synthesized from Snell Shells using a slightly modified method by Gumel *et al.* (2018). This method crushed the shells to reduce their size to 150 $\mu$ m mesh. The crystal samples were subjected to the following processes.

#### *Demineralization process*

The substrates were treated with 5% HCl w/v (1:10) for 30 Minutes at 30°C to remove the mineral content, mainly CaCO<sub>3</sub>. The pasty matter was washed under running tap water, rinsed with distilled water, and dried in a hot oven at 50°C for 24 hrs. The resultant Chitinoprotiec complex was subjected to the deproteinization process as in equation 1:



Equation 1

*Deproteinization process*

The sample was treated with 5% NaOH (w/v, ML: ratio = 1:10) for 30 minutes at 30°C with constant stirring for homogeneity. The resultant chitin was washed, dried, and milled into powder with an electrical blender. The reaction is shown in Figure 3. The resultant chitin was subjected to the deacetylation process

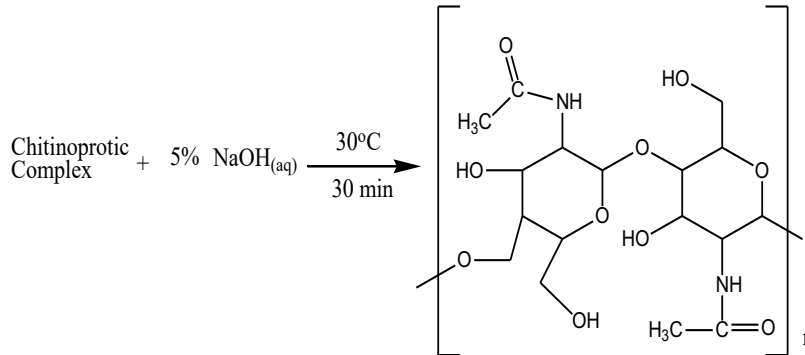
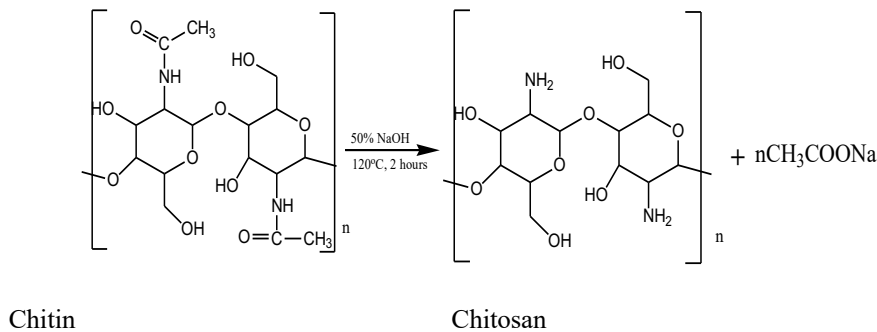


Fig. 3: De-proteinization process

*Deacetylation process*

The chitin sample was subjected to alkaline treatment with 50% NaOH solution (w/v) M:L ratio 1:20) for 2 hours at 120°C with periodic stirring for homogeneity. The resultant chitosan was kept neutral and oven-dried at 50°C for 24 hours. The chitosan obtained was milled into fine powder and kept in an airtight container. The reaction Scheme is shown in Figure 4.



Chitin

Chitosan

Fig. 4: Deacetylation of chitin

## Synthesis of Polylactic Acid from potato waste starch

### Extraction of potato peel starch

To prepare sweet potato peel starch, 500g of the potato peel was crushed and powdered to obtain the fine powder by sieving through a 250 $\mu$ m mesh. Two hundred grams of the fine powder was mixed with 600ml of water and stirred for 10 minutes to obtain the starch suspension. The mixture was allowed to settle for 3 hours. Then, the fine starch was then separated from the liquid by gently decanting the water part. The solid part was then oven-dried at 50°C for 72 hours. The dried fine powder was stored in an airtight container before use.

### Test for glucose

Test for glucose content was carried out using a slightly modified method by Suryani *et al.* (2017). In this process, 1 g of each starch powder was dissolved in 2 ml of distilled water in two test tubes. To the starch suspension, 1 ml of Fehling solutions I and II were added to each test tube, and the mixtures were shaken for 1 minute and then heated; the temperature was raised to boil. Brick red precipitates were observed in each test tube, confirming glucose in the starch.

## Synthesis of Polylactic Acid

The synthesis of polylactic acid is summarized in Figure 5. A hundred grams of the potato starch was hydrolyzed by adding 5% HCl. The mixture was heated on a hot plate at a temperature 60°C with constant stirring for 3 hours. The glucose was then inoculated with lactobacillus bulgaricus and incubated in an orbital shaker for 3-days to produce lactic acid (Akbar *et al.*, 2019). The lactic acid was then polymerized to form polylactic acid.

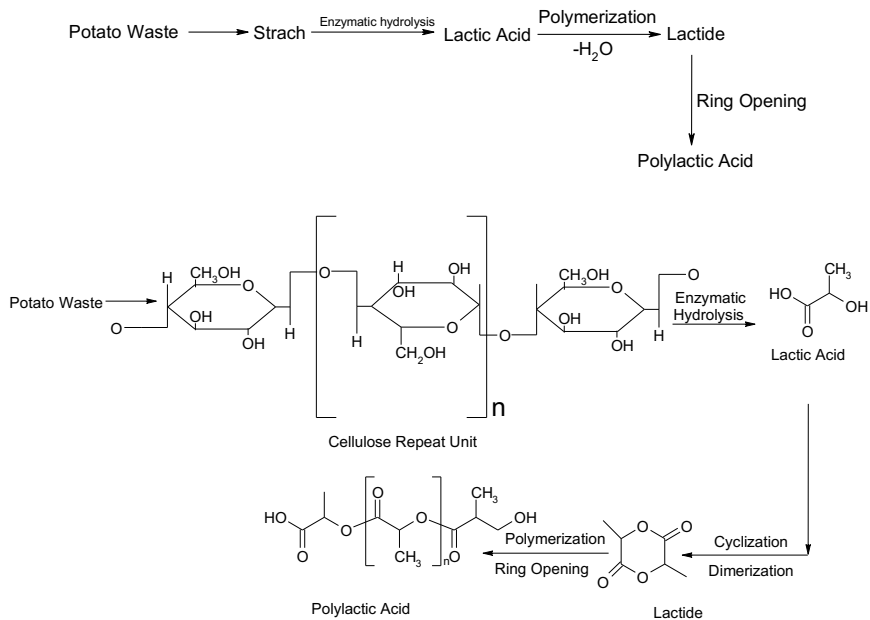


Fig. 5: Schematic representation of polylactic acid synthesis

### Modification of polylactic acid with chitosan

Poly (lactic acid) modification was carried out by an oxidation reaction between the amino groups in the glucosamine unit of chitosan and the terminal carboxyl group of polylactic acids chains, as summarized in Figure 6 (Suryani *et al.*, 2017). Two grams of chitosan powder was added to the polylactic acid, and the mixture was dissolved in 200ml of THF. The two solutions were mixed and stirred at 350rpm at 30°C for 3 hours (Formin *et al.*, 2017). The modified polylactic acids were purified by dialysis in deionized water for 48hours with water changes at 6hrs intervals, after which the polylactic acids were recovered and freeze-dried for 48hrs under a vacuum atmosphere of 0.5 bar at -46°C in the collector (Layla *et al.*, 2018). The process was repeated using 4g and 6g of chitosan.

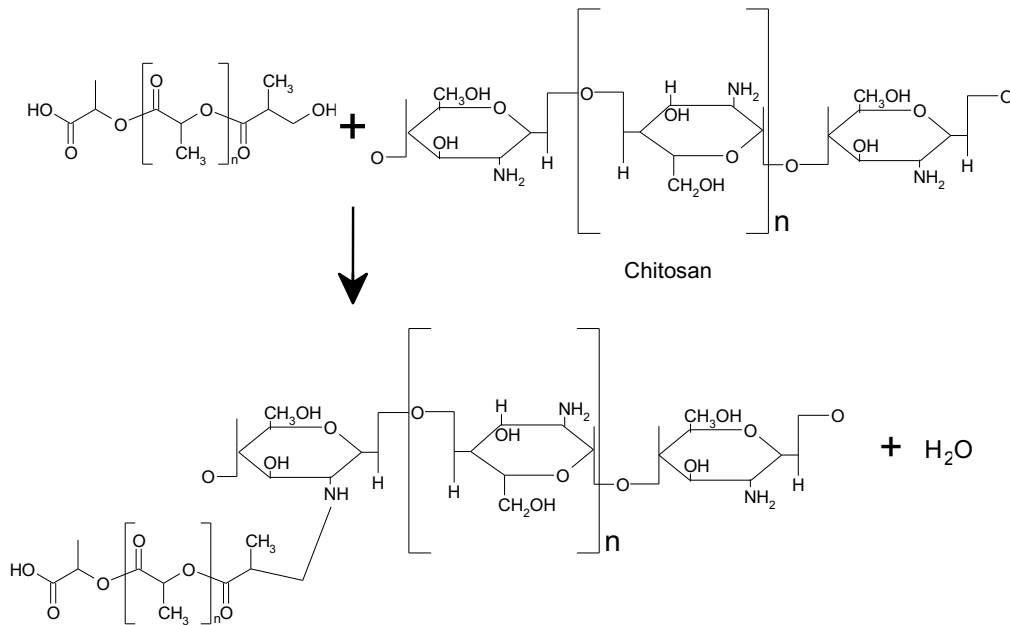


Fig. 6: Schematic representation of modification of polylactic acid with chitosan

### Extraction of jute fibers

Jute Fibres were extracted from the Jute plant using the slightly modified method by Shehu *et al.* (2017). In this process, the Jute stalks were soaked in enough water and allowed to stand for two weeks to enable the growth of Microorganisms (Bacteria and Fungi) that will break down the soft tissues from the stalks, leaving the cellulose intact. The stalks were removed, and the Fibres were washed and dried.

### Synthesis of thermoplastic film composites of chitosan-modified polylactic acid reinforced with jute fiber

The chitosan-modified polylactic acid-reinforced jute fiber film was prepared using 1%, 2%, 3%, 4%, and 5% Jute fiber. Green Thermoplastic Composites were prepared using the formulations in Table 1. Green thermoplastic reinforced with Jute fibers at the different masses of the filler was synthesized using the method of Nurshahida *et al.* (2018) with a slight Modification.

Table 1: Formulation of ingredients to produce green thermoplastics

Material	Treatment				
	A	B	C	D	E
Chitosan solution in 1% acetic acid	0%	5%	10%	15%	20%
Potato peel starch (g)	10	10	10	10	10
Jute fiber (g)	1	2	3	4	5
Distilled water (ml)	100	100	100	100	100

Each film used Potato peel starch as the matrix at a constant mass of 10g. The acetic acid (1%) was used to dissolve the chitosan at an M:L ratio of (1:10) to form the chitosan solution, while the Jute fiber was used as a filler in the film form reaction at (1,2, 3, 4 & 5) g per batch of the preparation. Hence, the green thermoplastic films were prepared by measuring 100ml of distilled water in a 1-litre beaker, 10g of the potato peel powder was added to the beaker, and the mixture was stirred with a glass rod. The mixture was heated to about 120°C until the gelation of the starch was achieved. The starch solution was allowed to cool; 5% of the chitosan solution was added to the mixture and stirred. Five milliliters of 0.1M HCl were added to the mixture and stirred with a glass rod. The mixture was heated at 70°C for 15min until the mixture became transparent. 1g of long Jute fibers was laid on the casting mold lined with foil paper. The mixture was poured into a mold containing the Jute sheath and allowed to cool at room temperature for 3 days. The thermoplastic film Jute composite was then baked at 18°C for 15 minutes and allowed to cool at room temperature for 24 hours. The thermoplastic film was removed from the aluminum foil and packed in a glass container. The procedure was repeated using CHT 10, 15, 20%, and jute fiber 2g, 3g, 4g, and 5 for each film. Thus, 12 samples were prepared and characterized by the following.

### Characterization

The films were characterized using FTIR, SEM, and mechanical properties using tensile test equipment, a universal testing machine (model exceeding E. 43, Japan), and Enzymatic degradation to assess biodegradability Vinceto, (2018).

## RESULTS AND DISCUSSION

### FTIR Spectrum

From Table 2 and Figure 7 (a,b,c), it was established that absorption bands in the region of 3000-2800cm<sup>-1</sup> were due to OH stretching and bending vibration in both chitosan, PLA, and PLA-Ch composites. The absorption peaks at 2800-3000cm<sup>-1</sup> were due to CH stretching and bending vibration in the glucose pyranose rings of chitosan, PLA, and Ch-PLA Nanocomposites. At the same time, absorption bands at 1750-1760C were due to C=O stretching vibrations in chitosan, PLA, and the Ch-PL matrix. Another peak at 1428 – 1448 was due to CH-Bending vibration. Absorption at 1554cm<sup>-1</sup> was due to NH stretching vibration in the 2<sup>o</sup> amide confirming amidation reaction.

Table 2: The FTIR spectrum of chitosan, modified polylactic acid, displayed the following absorption characteristics.

Sample	Absorption band (cm <sup>-1</sup> )	Functional group
Chitosan	300 – 3600	OH stretching bending vibration
	3457	NH stretching and bending
	1659 – 1759	C = O stretching
	1547	NH <sub>2</sub> stretching
	2918,2873	CH <sub>2</sub> stretching
PLA	3365	OH stretching vibration
	1760	C = O stretching and bending vibration in the amide
	2800 – 3000	CH – Stretching and bending vibration in the glucopyranose ring.
		CH Stretching and wagging
PLA-Ch	3450	OH stretching
	1554	NH – Stretching in the 2 <sup>o</sup> amide confirming Amidation reaction
	2840	CH-Stretching

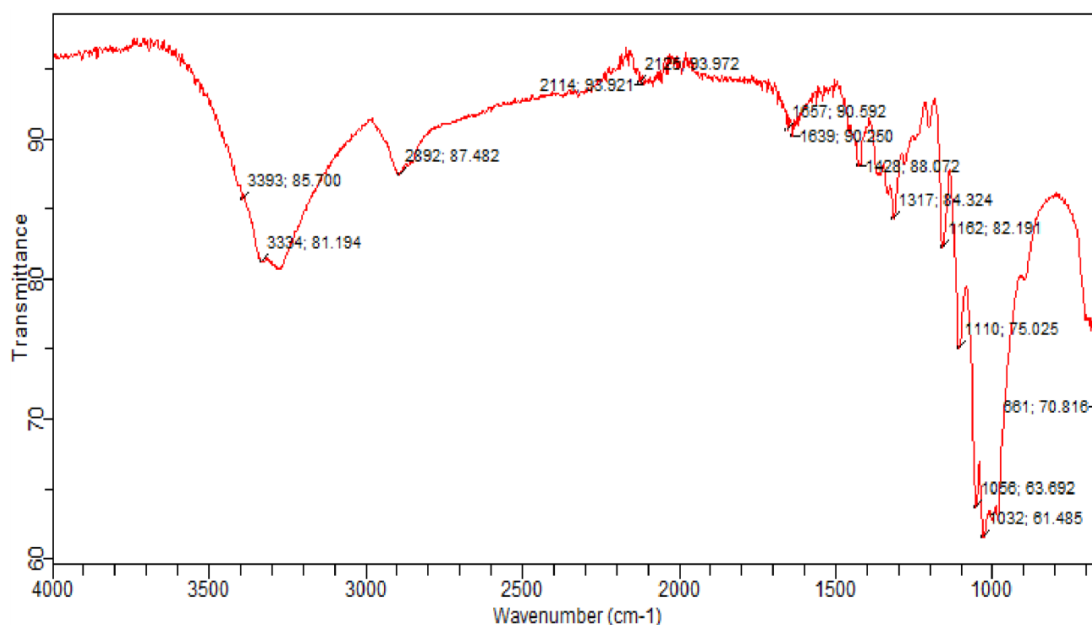


Figure 7a: FTIR spectrum of poly(lactic acid)



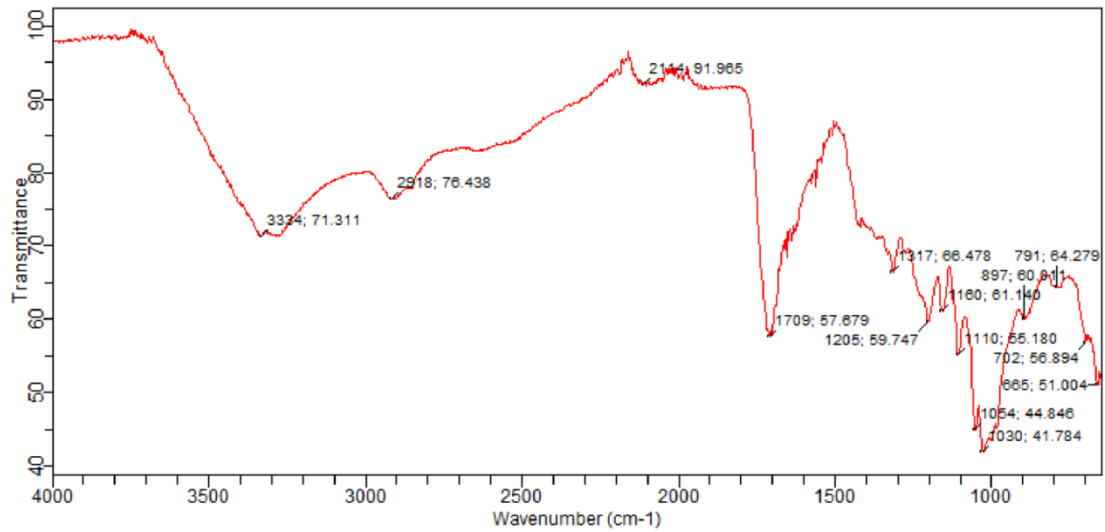


Figure 7b: FTIR Spectrum of Chitosan

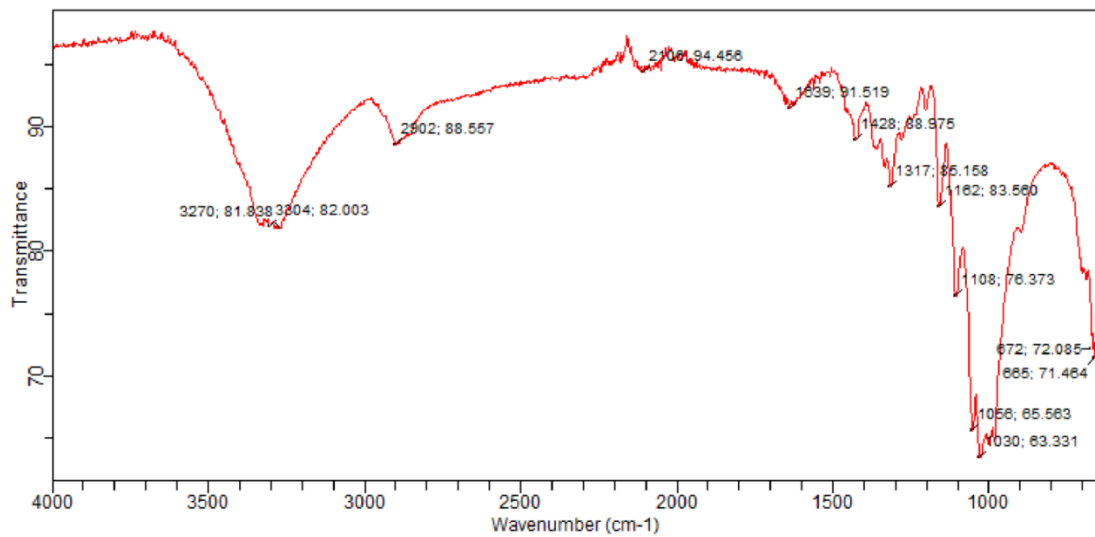


Figure 7c: FTIR Spectrum of Chitosan modified Poly(lactic acid)

### Mechanical properties of jute fiber/PLA-Ch composite film

Analysis of tensile strength and elongation at the break of the composite films yield very good results. The results are depicted in table 3.

Table 3: Tensile testing values for Ch-PL/jute fiber composites

Jute-Fiber (wt%)	Sample volumes 100ml & 200ml	Tensile strength (PMA)	Elongation (%)
5	Ch-PLA (wt%) 95 in 100ml	140.960	32.38
10	90 in 100ml	186.200	31.96
15	85 in 100ml	200.610	30.19

From Table 3, it was observed that the tensile strength of the composites increases as a factor of polylactic-Chitosan ratio, which indicates that the higher the increases in PLA-Ch Concentration (%), the higher the tensile strength and stiffer the composites films. Conversely, the elongation to break off the composites decreases with increased concentration of PLA-Ch.

### Scanning Electron Microscopy of the Bioplastics

The Ch-PLA/jute fiber bioplastics morphology showed a homogeneous surface independent of the concentration of Ch-PLA or the amount of jute fiber in the composites. The results may be attributed to the Ch-PLA's biocompatibility with the composites' jute fiber. The results conform to the findings of Nurshahida and Zainoha (2016). The micrographs of Ch-PLA, Jute/Ch-PLA, and chitosan are shown in Figure 8.

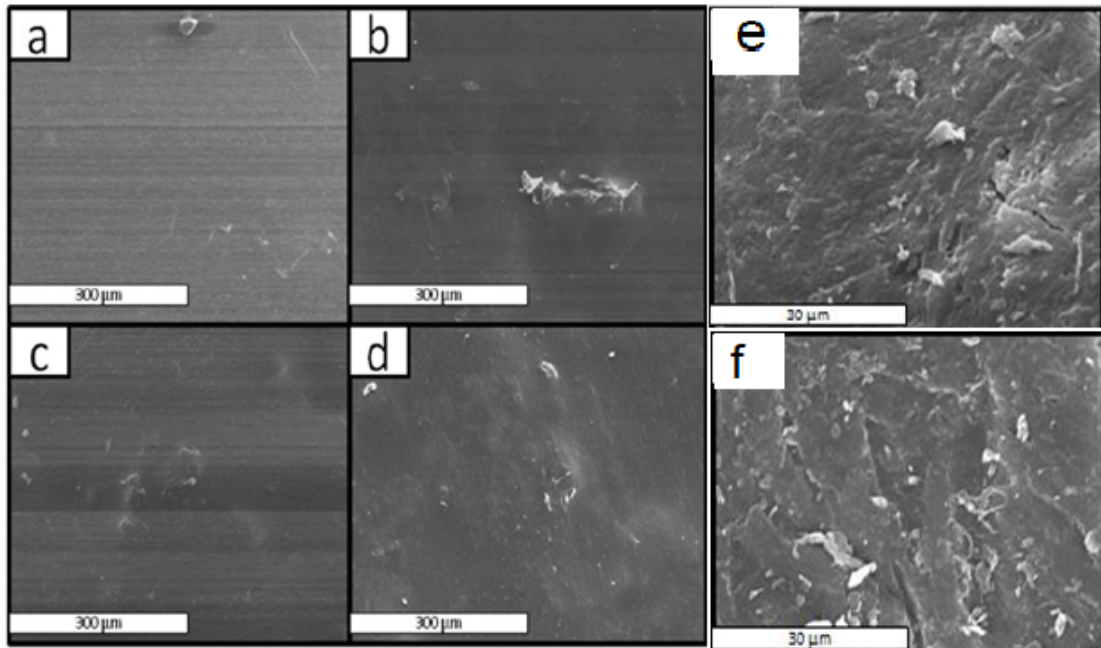


Figure 8: (a) Chitosan (b) PLA (c) Ch-PLA (d) 5% jute /Ch-PLA (e) 10% jute /Ch-PLA (f) 15% jute /Ch-PLA

## **CONCLUSION**

The escalating environmental issues linked to petrochemical-based plastics underscore the urgent need to develop sustainable alternatives. This review examines the viability of utilizing potato waste starch as a renewable feedstock for bioplastic production, emphasizing its considerable environmental and economic benefits. Incorporating chitosan as a modifier within the polymer matrix enhances the bioplastic's antimicrobial, antifungal, and antiviral properties and improves its biocompatibility, bioadhesiveness, and non-toxicity. Additionally, the reinforcement of the bioplastic with jute fiber markedly augments its mechanical properties, rendering it suitable for packaging applications. Characterization studies conducted through Fourier Transform Infrared Spectroscopy (FTIR) and tensile testing indicate that increased chitosan concentration correlates with improved tensile strength, albeit with a concurrent decrease in elongation at break. These findings elucidate the feasibility of producing environmentally friendly, high-performance bioplastics derived from waste materials, thereby presenting a promising approach to mitigating plastic pollution while simultaneously contributing to the advancement of a circular economy.

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## **AUTHOR'S CONTRIBUTION**

All the authors equally contributed to writing, reviewing, approving corrections, and approving the publication submission.

## **CONFLICT OF INTEREST STATEMENT**

The authors affirm that there are no competing interests regarding the publication of this paper.

## **REFERENCES**