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Evaluation of Tensile Strength and Elongation of Bioplastic Films Manufactured from Cellulose of Local Amber Rice Husks Extracted by Chemical, Physical, and Biological Treatments

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Abstract

The use of bioplastics can be a promising option to achieve environmental sustainability and reduce environmental pollution, as it has a positive impact on reducing climate change and reducing greenhouse gas emissions compared to traditional plastics made from petrochemicals, which cause environmental pollution due to increased carbon dioxide emissions into the atmosphere as a result of burning and destruction of conventional plastics, which causes in major climate change due to rising temperatures and the occurrence of the global warming phenomenon. The high cellulose contents in Amber Rice Husk (ARH) support use as raw material for bioplastic synthesis and include the following components, cellulose 36.48%, hemicellulose 39.5%, lignin 7.14%, protein 1.09%, and ash 18.91%. This study aims to prepare a bioplastic film from chitosan, glycerin, sorbitol, and cellulose extracted from ARH using three chemical, physical, and biological processes, in addition to the measurement of the physical properties, such as tensile strength, and elongation. Three treatments (chemical, physical, and biological) were used in cellulose extraction from ARH to synthesize bioplastic films. The best treatment for cellulose production was the physical at a concentration of 61.05%, while chemical and biological treatments were 45.62, and 45.06%, respectively. The study primarily focuses on mechanical properties, such as tensile strength and elongation, essential for evaluating bioplastic usability in real-world applications. The result of tensile strength for the optimum bioplastic film, manufactured by chitosan (0.6, 0.8, and 1 g) and glycerol (2.0, 2.5, and 1.5 ml) added to the cellulose (0.8, 0.8, and 1 g) extracted by chemical, physical and biological treatments of the ARH respectively, proven that chemical and biological treatments (0.566 and 0.655 N/mm^2), respectively, were slightly lower than physical treatment with (0.754 N/mm^2), whereas, the elongation percentage of bioplastic film prepared by physical treatment was recorded at 20.44% lower than biological and chemical treatments at 28.66 and 24.42%, respectively.

Keywords: Agricultural waste, bioplastic, mechanical properties, sustainable environment. Amber Rice Husk (ARH), cellulose.

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تقييم قوة الشد والاستطالة لافلام البلاستك الحيوي المصنع من سليلوز قشور رز العنبر المحلي المستخلص بالمعاملات الكيمياوية والفيزياوية والبايولوجية

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الخلاصة

يمكن أن يكون استخدام البلاستيك الحيوي خيارا واعدا لتحقيق الاستدامة البيئية والحد من التلوث البيئي، حيث أن له تأثير إيجابي على الحد من تغير المناخ وتقليل انبعاثات الغازات الدفيئة مقارنة بالبلاستيك التقليدي المصنوع من البتروكيماويات والذي يسبب التلوث البيئي بسبب زيادة ثاني أكسيد الكربون والانبعاثات إلى الغلاف الجوي نتيجة حرق وتدمير المواد البلاستيكية التقليدية، مما يتسبب في تغير مناخي كبير يسبب ارتفاع درجات الحرارة وحدوث ظاهرة الاحتباس الحراري. تدعم محتويات السليلوز العالية في قشور رز العنبر (ARH) استخدامها كمادة خام لتخليق البلاستيك الحيوي وتشمل المكونات التالية، السليلوز 36.48%، الهيمسيلولوز 39.5%، اللجنين 7.14%، البروتين 1.09%، والرماد 18.91%. تهدف هذه الدراسة إلى تحضير فيلم بلاستيكي حيوي من مادة الكايتوسان، الكلسرين، السوربيتول، والسليلوز المستخلص من ARH باستخدام ثلاث معاملات كيميائية، فيزيائية، وبيولوجية، ومن ثم قياس الخواص الفيزيائية، مثل قوة الشد، والاستطالة. تم استخدام ثلاث معاملات (كيميائية وفيزيائية وبيولوجية) في استخلاص السليلوز من ARH لتصنيع أفلام البلاستيك الحيوي. وكانت أفضل معاملة لإنتاج السليولوز هي المعالجة الفيزيائية بتركيز 61.05%، بينما بلغت المعاملات الكيميائية والبيولوجية 45.62، و45.06% على التوالي. تركز الدراسة في المقام الأول على الخواص الميكانيكية، مثل قوة الشد والاستطالة، وهي ضرورية لتقييم قابلية استخدام البلاستيك الحيوي في تطبيقات العالم الحقيقي. نتيجة قوة الشد لفيلم البلاستيك الحيوي المصنع من الكايتوسان (0.6، 0.8، 1 غم) والكلسرين (2.0، 2.5، 1.5 مل) مضافًا إلى السليلوز (0.8، 0.8، 1 غم) المستخلص كيميائيًا وفيزيائيا وبايولوجيا على التوالي. أثبتت أن المعاملات الكيميائية والبيولوجية (0.566 و 0.655 نيوتن / ملم^2)، على التوالي، كانت أقل قليلاً مقارنة بالمعاملة الفيزيائية (0.754 نيوتن / ملم^2)، بينما سجلت الاستطالة المئوية للبلاستيك الحيوي المحضر بالمعاملة الفيزيائية بنسبة 20.44% أقل من المعاملات البيولوجية والكيميائية عند 28.66 و24.42% على التوالي.

Introduction

Plastic is a crucial substance used by humans for food packaging. Because it takes thousands of years to degrade, it can collect and cause pollution and environmental issues [1]. To overcome this property, it is possible to use packaging as a sustainable alternative, whether or bio-based materials in their composition. According to the reference [2], this type of biodegradable plastic is susceptible to rapid breakdown but has low mechanical strength.

The increasing accumulation of environmental pollutants, especially agricultural waste, in the environment, the combustion of which exacerbates the global energy crisis and the emission of greenhouse gases, which are considered one of the factors contributing to the phenomenon of global warming [3], has led to thinking about extracting the necessary materials as sources in the bioplastics industry, such as cellulose, hemicellulose, and lignin, which are found in the organic part of agricultural waste, in addition to considering them a carbon source for the production of the cellulase enzyme when microorganisms grow and reproduce on them [4]. The content of cellulose in different agricultural waste is as follows:

61.8% in rice straw [5], 40% in sugarcane bagasse [6], and 17.5% in tea waste [7]. This cellulose can produce bioplastics, which can help solve waste-related environmental issues and increase their useful life.

One of the methods of bioremediation of conventional plastics is recycling polymers to improve the performance of environmentally friendly recycled materials and overcome the problems caused by conventional plastics, especially synthetic plastics such as polystyrene, polyethylene, and ethylene vinyl acetate, which some insects, such as flour beetle and *Galleria mellonella* larvae digest [8,9], in addition to using microorganisms, such as bacteria and fungi extracted from soil, artificial clay, compost, sludge, and water, to decompose hydrocarbon waste and control plastic accumulation while using it as a carbon source [10,11], and reducing the consumption of fossil fuel-based chemicals; however, the quality of recycled polymer products is due to chemical contaminants present during plastic waste recycling [12]. Moreover, these procedures include polymer classification, washing, grinding, and extrusion, which will limit the plastic recycling cycle and cause some degradation [13].

The production of bioplastics depends on several materials, such as sugars (cellulose, starch, pectin, chitin), proteins (gluten, casein, gelatin), animal fats, vegetable oils, or materials produced by microalgae, as these materials can be biodegraded without negatively affecting the environment and consuming no energy [14]. On this basis, we focused our research on the exploitation of agricultural waste in the local environment, such as ARH, because it's the common local Iraqi product rice husks contain cellulose as one of their basic components and are considered a suitable material to replace synthetic polymers in the production of bioplastics [15].

In contrast to conventional polymers, which take 500-1000 years to degrade, bioplastic biodegradable polymers deteriorate when exposed to environmental factors such as temperature, humidity, oxygen, and UV light. Bioplastics combine conventional and biodegradable polymers, enabling controlled biodegradation [16]. Bioplastics are environmentally sensitive, they can be fully degradable or recyclable, enabling industry recovery and recycling processes. Bioplastics and sustainable materials are crucial for a more sustainable world. Development in material science has the vision to avoid such detrimental environmental effects, which were common in the past century, such as dumping large amounts of waste into oceans and air pollution [17]. The sustainability challenge is an ecological-economic opportunity that forces the research community to go ‘beyond the generic’ in terms of the material characteristics of sustainable materials [18].

Tensile strengths for conventional plastics and bioplastics range from 24-302 MPa and 10-100 MPa, respectively, according to standard specifications; however, most starch-based bioplastics and cellulose acetate butyrate have tensile strengths that fall outside of this range [19]. A polymer's tensile strength depends on its molecular weight, degree of crosslinking, and crystallinity. Thus, the tensile strength and other mechanical characteristics of plastics are determined by the chemical structure of the raw material and the physical or chemical structure of the polymer [20].

Therefore, to overcome their weakness and enhance their flexibility, workability, and brittleness when they are equilibrated at ambient relative humidity, plasticizers like glycerol, which form bonds with water molecules, are commonly added to starch-based bioplastics during the bioplastic synthesis process [21].

A filler made of cellulosic materials was required to boost the mechanical strength of bioplastic [22]. This improves thermoplastic starch's biocompatibility, renewability, and sustainability while decreasing its hydrophilic. These factors make bioplastics more environmentally friendly than polymers made from fossil fuels.

This study aims to manufacture a bioplastic film from chitosan, glycerin, sorbitol, and cellulose extracted by using different treatments of ARH, such as chemical, physical, and biological, and then evaluates the physical properties of bioplastic films, such as tensile strength and elongation percentage.

Materials and Methods

Collection of ARH

The ARH was collected from agricultural waste disposal sites in the Iraqi provinces, located in Kufa, Najaf Province, and Al-Kifl, Babel Province, washed, and dried for one hour at 60°C in an oven, sieved by an 80-mesh sieve, and then stored in bags to facilitate its continued use for cellulose extraction [23].

Chemical extraction

Cellulose was extracted by the de-lignification process, which occurs via treating ARH (50 g) with 12% (500 ml) sodium hydroxide (NaOH) and heating to 80°C for 3 h, then washed with distilled water until it reached a neutral level. The remaining RH was heated for an hour at 80°C after being treated for bleaching with a 2.5% sodium hypochlorite (NaClO) solution [24]. Therefore, the residue was filtered and cleaned several times with distilled water and dried at 60°C in an oven until it achieved a constant weight (Figure 1) [23].

Physical extraction

To perform the physical extraction of cellulose, 50 g of ARH was added to 500 ml of distilled water and autoclaved for 30 min at 121°C. Then, according to reference [22], ARH was filtered to bleach for one hour at 80°C using a 2.5% NaClO solution, washed multiple times to achieve a neutral pH, and dried at 60°C until it reached a constant weight (Figure 1).

Biological extraction

Fifty grams of dried ARH were treated for cellulose extraction with 500 ml of crude garbage enzyme (25%), extracted from the fermentation process of food waste, and incubated for 7 days at room temperature in a shaker incubator, then washed twice with distilled water, and next bleached with 500 ml NaClO (2.5%) solution agent and heated for an hour at 80°C. Finally, the residue was washed until it reached a neutral pH value and dried in an oven at 60°C to a constant weight (Figure 1) [25].

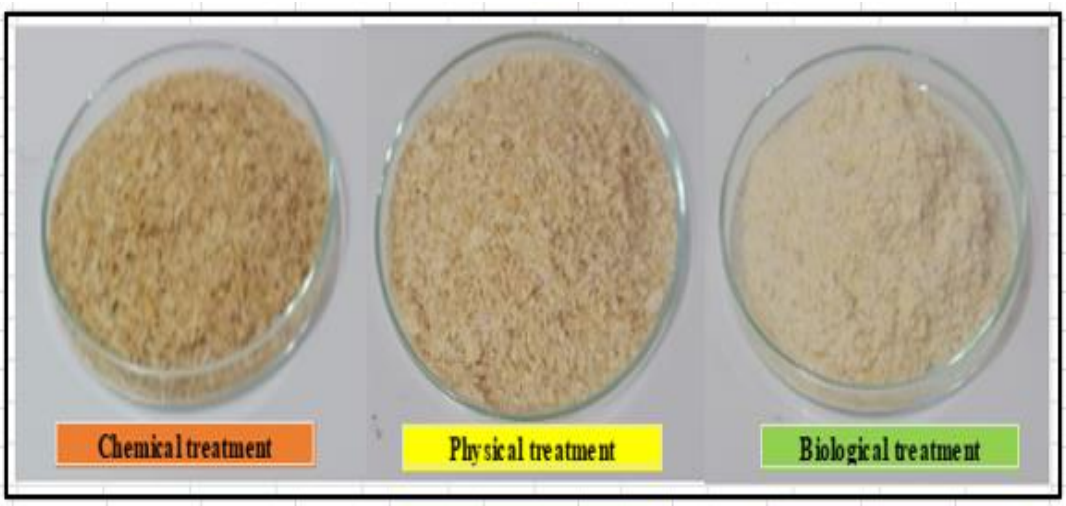

Figure 1: Different extraction treatments of cellulose of ARH

Optimum condition of bioplastic film preparation

Bioplastic was prepared by mixing cellulose and chitosan (0.2, 0.4, 0.6, 0.8, and 1 g), (0.2, 0.4, 0.6, 0.8, and 1 g) and dissolving the mixture in 50 ml of 1% acetic acid. Then add 0.5 g of sorbitol and glycerol weights (0.5, 1, 1.5, 2.0, and 2.5 ml) as a plasticizer, the mixture was mixed by heating it at a temperature of 50°C for one hour and poured into glass dishes until solidified (Figure 2) [26].

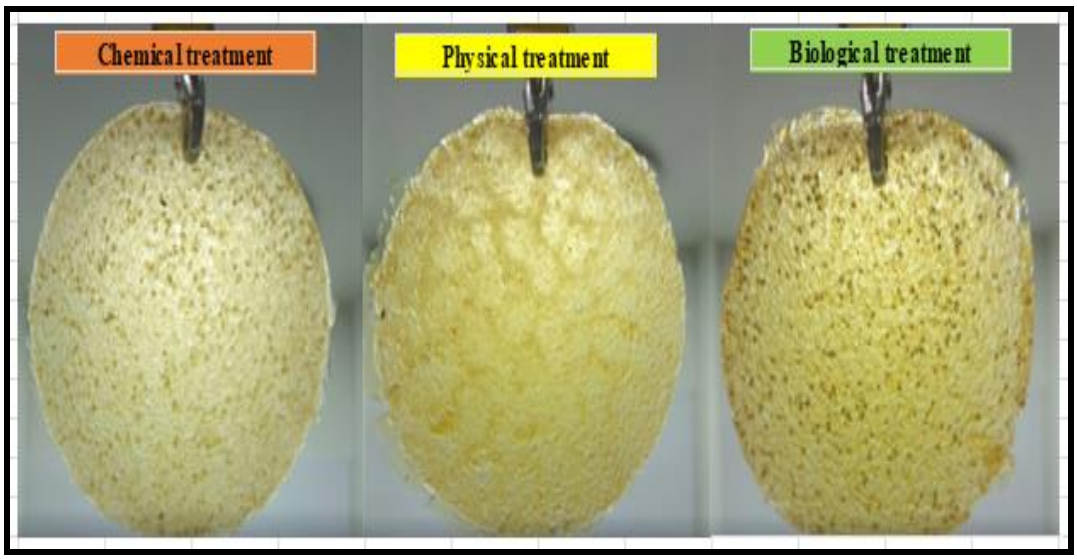

Figure 2: Bioplastic synthesis by different extraction treatments of cellulose from ARH

Then, the tensile strength and elongation of the prepared bioplastic films were measured, and the biofilm characterized by ideal tensile strength and elongation was extracted, and its components of cellulose, chitosan, and glycerol were recorded as optimal components.

Chemical composition of RH

A fiber analyzer device determines the chemical composition of the rice husk (RH) cell walls and the cellulose percentage of ARH after three chemical, physical, and biological treatments.

Measurement of the mechanical properties of bioplastic films

The manufacture of bioplastic films was separated into strips to determine their physical characteristics [27].

Tensile strength

Tensile strength test to bioplastic film was done with (Tinius Olsen device H50KT / England) which is commonly measured in mega Pascal (MPa) units. Every bioplastic strip was stretched to its breaking point. Tensile strength testing aims to assess a bioplastic strip's ability to withstand loading at the bending point and to look at its elasticity at the rupture point, as in equation 1 [28].

$$\text{Equation 1:} \quad TS = F_{max} / A_0$$

TS = tensile strength
F_{max} = maximum force
A_0 = initial surface width

Elongation

According to equation 2, elongation is the percentage change in the length of the bioplastic strip when the film stretches until it separates. To obtain maximum tension in each strip to stretch or elongate, the measurement of tensile strength at break aids in estimating the amount of force required [29].

$$\text{Elongation (\%)} = (\text{Change in length} / \text{Original length}) \times 100$$

$$\text{Equation 2:} \quad \text{Elongation (\%)} = [(L_1 - L_0) / L_0] \times 100$$

L_1= the final length of the test object
L_0= the initial length of the test object

Results and Discussion

Cellulose extraction

The chemical composition of the rice husk cell wall, as determined by a Fiber Analyzer Device, is displayed in Figure 3, and includes the following components for ARH cellulose 36.48%, hemicellulose 39.5%, lignin 7.14%, protein 1.09%, and ash 18.91%. Natural polysaccharide polymers and their derivatives have been the subject of studies of biodegradable bioplastics [30]. Compared to other polysaccharides, cellulose has been of interest in applications due to various advantageous properties, particularly low cost, high strength, biodegradability, lightweight, and sustainability [31]. Cellulose-based materials have been applied in many fields, such as food packaging [32], reinforcement materials [33], coating materials [34], wastewater treatment [35], biodegradable packaging, flexible optoelectronics, and lightweight automobiles [36], and was the most abundant biopolymer, is primarily derived from plants [37]. In addition, agricultural wastes have been used as secondary bioresources to extract cellulose [38], including sugarcane bagasse [39] and rice straw [40].

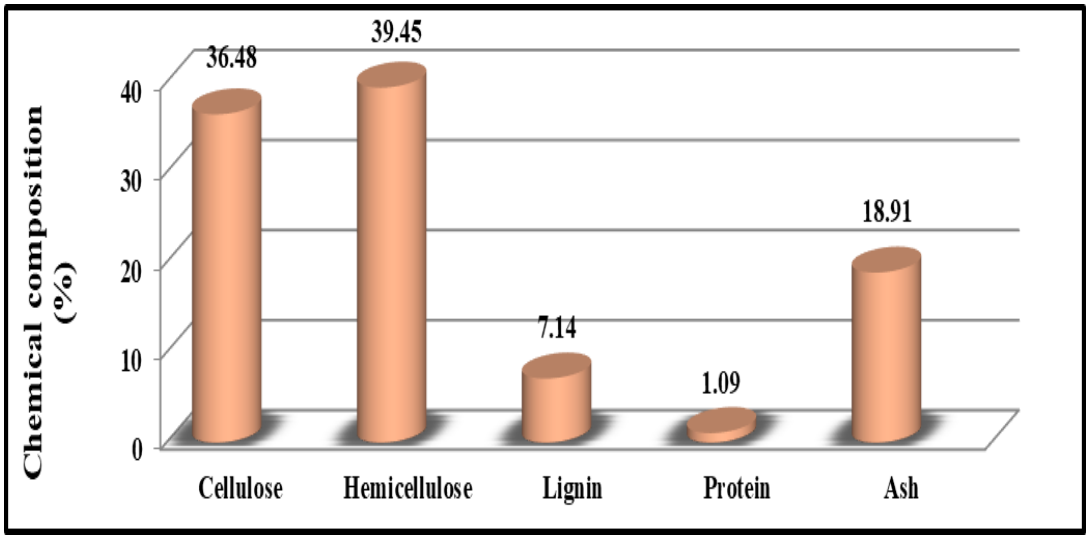

Figure 3: Chemical composition of ARH

The cellulose concentrations extracted from ARH by chemical, physical, and biological methods were 45.62, 61.05, and 45.06%, respectively (Figure 4).

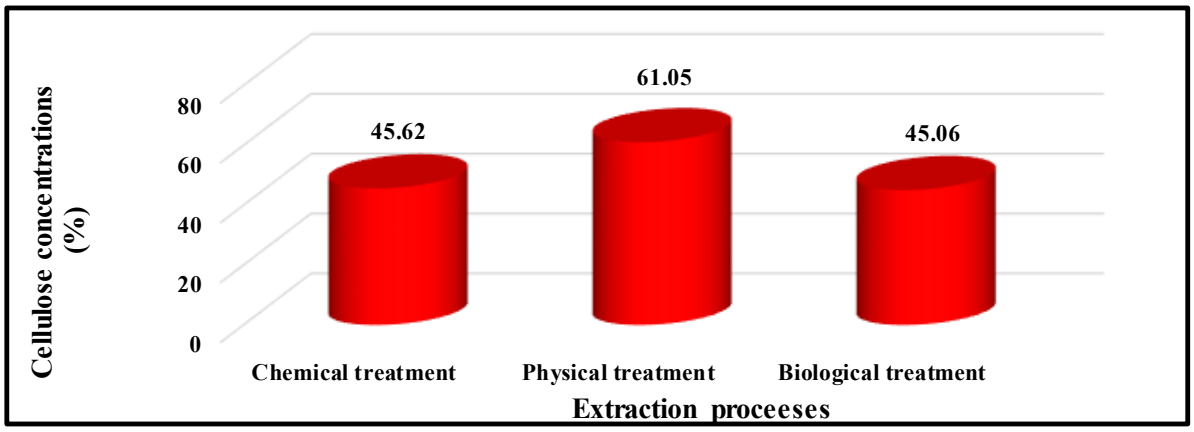

Figure 4: Percentage of cellulose extracted from ARH by chemical, physical, and biological extractions

Chemical extraction

Sodium hydroxide solution increases the yield of cellulose fiber during chemical extraction by breaking hydrogen bonds between the lignocellulosic components, which dissolve in alkali solutions to form black liquor [41]. After the alkali extraction, the lignin was removed using NaClO as a bleaching agent because NaOH alone was unable to remove all the lignin. NaClO contains a hypochlorite ion that can dissolve the ether bond in the lignin structure and improve the pulp's white brightness [42].

Strong hydrogen bonds in cellulose's structure prevented it from dissolving at a temperature below its degradation point [43]. Nevertheless, it was discovered that raising the NaOH concentration reduced the yield since some cellulose chains would break down during the extraction process at greater concentrations. Elevated concentrations of NaOH disrupt certain crystalline areas of cellulose, facilitating its easy dissolution in the solution treatment and consequently decreasing the cellulose fiber yield [44].

Physical extraction

Physical extraction by hydrothermal means is an easy and economical method. Hydrothermal pretreatment without chemical addition impacts the physical changes of cellulose by removing lignin on the surface, solubilizing hemicellulose, and improving the accessibility of the structure [45]. When cellulose is wet, water hydrogen bonds with the -OH groups in the cellulose's structure, causing the cellulose to be loosely packed. The chain becomes less mobile after increasing interchain bonds water from the cellulose, resulting in a shrinking and densely packed structure [46].

Biological extraction

Biological extraction involves soaking ARH in a 25% extract of raw garbage enzymes for a week, which is environmentally friendly because it doesn't require alkali or acidic materials like chemical treatment. However, the percentage of cellulose extracted by chemical and biological methods is similar: 45.62% and 45.06%, respectively.

The de-lignification and cellulose pulp formation processes may impact the chemical bonds that bind cellulose, hemicellulose, and lignin together. Conversely, the solvent eliminated or dissolved hemicellulose and lignin [47]. Experiments reported by Pramasari *et al.,*[48], and Bhatia *et al.,* [49], are about an alkaline pretreatment employing 3% NaOH coupled autoclaving at 121°C. Alkaline agents (NaOH) work on efficient pretreatment for de-lignification by rupturing hydroxyl ion (OH-) linkages between lignin and hemicellulose and dissolving the lignin polymer into its aromatic derivatives.

Tensile strength of optimum bioplastic film

Tensile strength is the ability of a bioplastic film to withstand a given load until the bioplastic breaks. Tensile strength for the bioplastic film (Figure 4) synthesis by chitosan (0.6, 0.8, and 1 g) and glycerol (2.0, 2.5, and 1.5 ml) added to the cellulose (0.8, 0.8, and 1 g) extracted from the chemical, physical and biological treatments of the ARH were (0.566, 0.754, and 0.655 N/mm^2) respectively.

The tensile strength of optimum bioplastic films prepared by chemical and biological treatments was slightly lower (0.566 and 0.655 N/mm^2), respectively, than physical treatment with (0.754 N/mm^2), results indicated interfacial adhesion forming a strong hydrogen bond network between cellulose, chitosan, and glycerol as plasticizers, as presented in Figure 5.

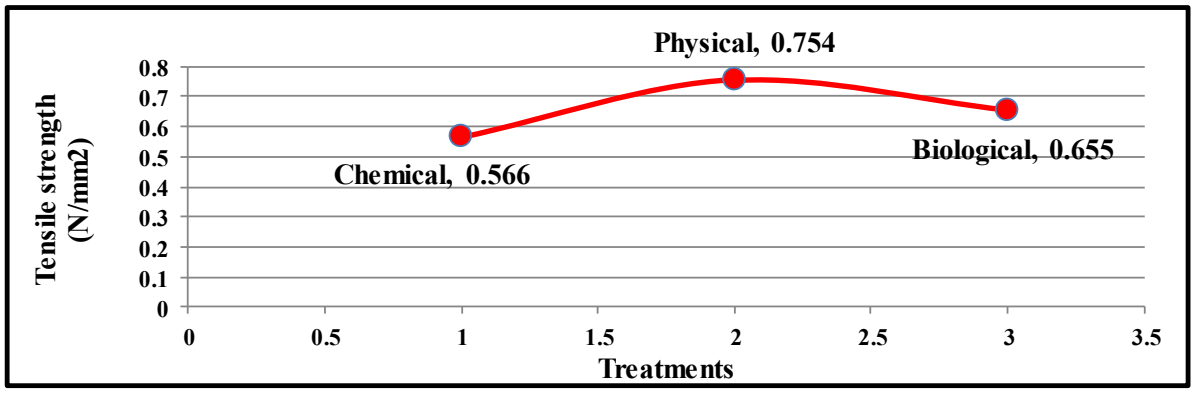

Figure 5: Tensile strength for bioplastic film synthesis by chitosan (0.6, 0.8, and 1 g) and glycerol (2.0, 2.5, and 1.5 ml) added to the cellulose (0.8, 0.8, and 1 g) extracted from the chemical, physical and biological treatments of the ARH

Tensile strength is one of the crucial fundamental characteristics needed for packaging materials, and it provides resistance to direct pull. The high value of the tensile strength of bioplastic stripes was attributed to the good interface adhesion and the formation of hydrogen bonds between cellulose and filler chitosan [50].

Elongation properties of optimum bioplastic film

Elongation is the appropriate stretch extension or increase in bioplastic film. It is a polymer's elasticity, flexibility, and tensile strength.

Elongation for bioplastic film synthesis by chitosan (0.6, 0.8, and 1 g) and glycerol (2.0, 2.5, and 1.5 ml) added to the cellulose (0.8, 0.8, and 1 g) extracted from the chemical, physical and biological treatments of the ARH were (24.42, 20.44, and 28.66%), respectively **(**Figure 6), based on different masses and volumes of cellulose, chitosan, and glycerol contents. These results indicate that the plastic film prepared by adding cellulose extracted by the physical method recorded less elongation than the chemical and biological methods, because the addition of cellulose to the bioplastic film may result in higher strength and lowered elongation, this may be due to hydrogen bonds between hydroxyl and carboxyl groups (COOH) of cellulose [21].

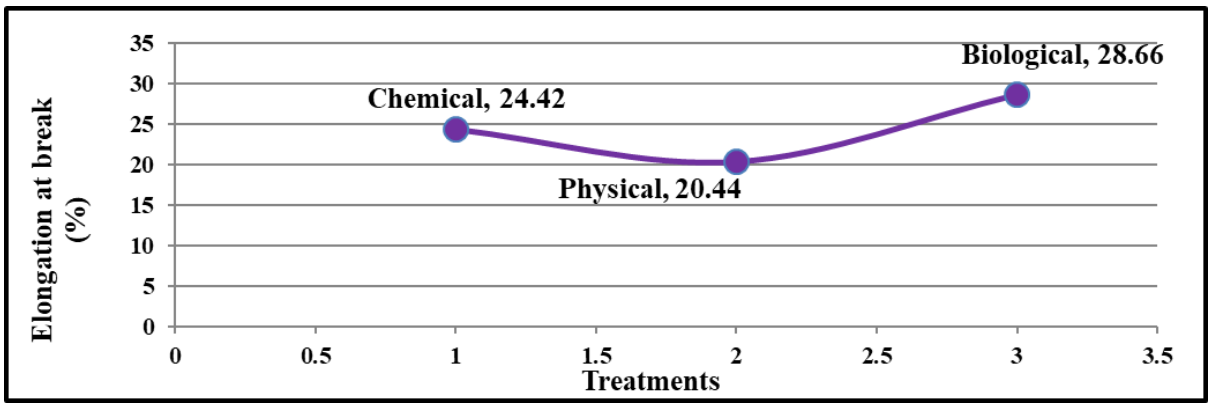

Figure 6: Elongation at the break for bioplastic film synthesis by chitosan (0.6, 0.8, and 1 g) and glycerol (2.0, 2.5, and 1.5 ml) added to the cellulose (0.8, 0.8, and 1 g) extracted from the chemical, physical and biological treatments of the ARH

Although chitosan will decrease the bioplastic's elongation due to its increased stiffness, adding more chitosan filler will also increase the hydrogen bonding interaction between the filler and the cellulose matrix, decreasing the bioplastic film's flexibility [51].

Conclusion

Compared to chemical and biological treatments, physical treatment of cellulose extraction from ARH is more economically viable and effective because it uses a hydrothermal process without chemicals. Additionally, a bioplastic made from cellulose combined with chitosan, glycerol, and sorbitol was found to have a higher tensile strength than chemical and biological treatments. The bioplastic stripes' maximal ability to withstand stretching forces without elongating before breaking is known as their tensile strength. Increasing the length with a tensile load as a percentage will cause a material with low tensile strength to break with greater elongation up to fracture.

Other critical mechanical properties, such as flexural strength, impact resistance, and modulus of elasticity, are essential for bioplastics usability in real-world applications. Also, a broader range of formulations or composite materials could reveal additional properties and

functionalities beneficial for various applications. Future research should focus on enhancing the performance and consistency of raw feedstock conversion into useful bioplastics. We are at a technological crossroads, transitioning from petrochemical-based plastics to bioplastics from renewable and non-renewable resources. This shift promises less dependence on fossil fuels and higher-value products like resins and chemicals for fully biodegradable and compostable artifacts.

Authors contribution statement

The Author E.H. contributed to implementing the manuscript plan steps, conducting some work-related transactions, analyzing the data, and contributing to the interpretation of the results and provided critical revisions, references of the manuscript preparation, and writing of the manuscript with support from the Author A. H. The Author H.S. contributed to the reviewing manuscript (supervisor). The Author A.H. contributed to developing the plan, practically supervising the research, preparing and providing research supplies of materials and laboratories, and reviewing and auditing the research plan as a co-supervisor.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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