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Review Article

Research on Cellulose Film in Degradable Wet Garbage Bags

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ISSN: 2770-6613

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Submission: August 11, 2022

Published: August 19, 2022

Volume 4 - Issue 1

How to cite this article: Li Yinxuan*. Research on Cellulose Film in Degradable Wet Garbage Bags. Polymer Sci peer Rev J. 4(1). PSPRJ. 000580. 2022.
DOI: 10.31031/PSPRJ.2022.04.000580

Abstract

Cellulose is a natural polymer compound with the largest reserves in nature and is considered to be the main raw material for energy and chemical industry in the future. However, the high degree of polymerization and high crystallinity of natural cellulose make it difficult to dissolve in conventional solvents, which greatly limits the application of cellulose. In recent years, a variety of new types of cellulose have been discovered, and this paper briefly introduces plant-derived cellulose materials. The cellulose-based composite film obtained by dissolving the cellulose solvent system has been improved to a certain extent in terms of strength and degradability and is expected to be applied to the problem of wet garbage bag breaking caused by garbage classification in recent years. At present, cellulose-based composite materials have been widely used in food preservation and antibacterial packaging materials, active packaging materials and high barrier packaging materials. Based on the latest research on cellulose membrane materials, this paper looks forward to the future development hotspots, aiming to provide references for cellulose dissolution and the development of new cellulose membrane materials.

Keywords: Cellulose; Biodegradability materials; Coating film

Introduction

In recent years, with the increasing popularity of garbage classification, as of June 2022, 281 cities across the country have begun to implement garbage classification [1]. Among them, the issue that citizens need to break the bag when throwing wet garbage has been controversial, so the research and promotion of biodegradable wet garbage bags have considerable development prospects. The team of Zheng Lirong, executive vice president of the New Rural Development Research Institute of Fudan University, has developed a coating and laminating process technology of biodegradable materials on paper bags made of fiber composites, which not only solves the waterproof function of garbage bags but also achieves rapid and complete degradation. So far, there is no research on using cellulose as a raw material to make films. Therefore, based on the existing research on cellulose films, this paper will focus on different extraction methods of cellulose, the preparation process and related properties of cellulose films, and their biodegradability expansion analysis and comparison.

As one of the most abundant natural polymer composite materials in the world, cellulose can produce nearly 100 billion tons of cellulose every year only through plant photosynthetic respiration [2]. It is widely found in wood, cotton and other natural plant raw materials. At the same time, marine organisms and algae and other microorganisms also contain a large number of such organisms. Among them, cellulose has many advantages such as natural greenness, easy biodegradation, non-toxicity and good reproducibility, and is a typical environment-friendly material. If the cellulose in plants can be utilized, it is of great significance to effectively alleviate the problem of global resource shortage and to solve the problems affecting the global energy and ecological environment. In this study, cellulose extracts from the residues of bamboo, jute, salix, sugarcane residue and corn starch-based composites were selected as the main raw materials for the preparation of degradable cellulose composite films, and a general introduction of cellulose The technical research results and technological progress of composite membranes, the future research direction is prospected and guessed, and it is expected to provide references for the technical research of degradable and functional composite cellulose membranes and the development of membrane technology in China.

Preparation of cellulose film

Extraction and dissolution of cellulose

Cellulose is a linear polymer, which is formed by β-D-glucose group by removing a molecule of water between the hydroxyl groups on the para position of carbon atoms. Since there are hydroxyl groups on each structural unit, it is in the fiber. Hydrogen bonds may be formed in the molecules of cellulose macromolecules, so crystalline structures will be formed between cellulose molecules. Due to this crystalline structure, cellulose is difficult to dissolve in general solvents. Currently, common cellulose dissolving systems include sodium hydroxide solution, nitroethanol solution, sodium chlorate solution and ionic liquid [3].

Extraction of cellulose from bamboo harvesting residues: Li [4] and other teams chose to wash the powder of bamboo harvesting residues (mainly including bamboo branches and bamboo parts) to remove impurities in the powder. Then, the washed and dried powder is reacted with sodium hydroxide to remove hemicellulose, lignin and other components in the bamboo to obtain alkaline cellulose. Finally, sodium Carboxymethylcellulose (CMC) is obtained by etherification and carboxymethylation (Figure 1).

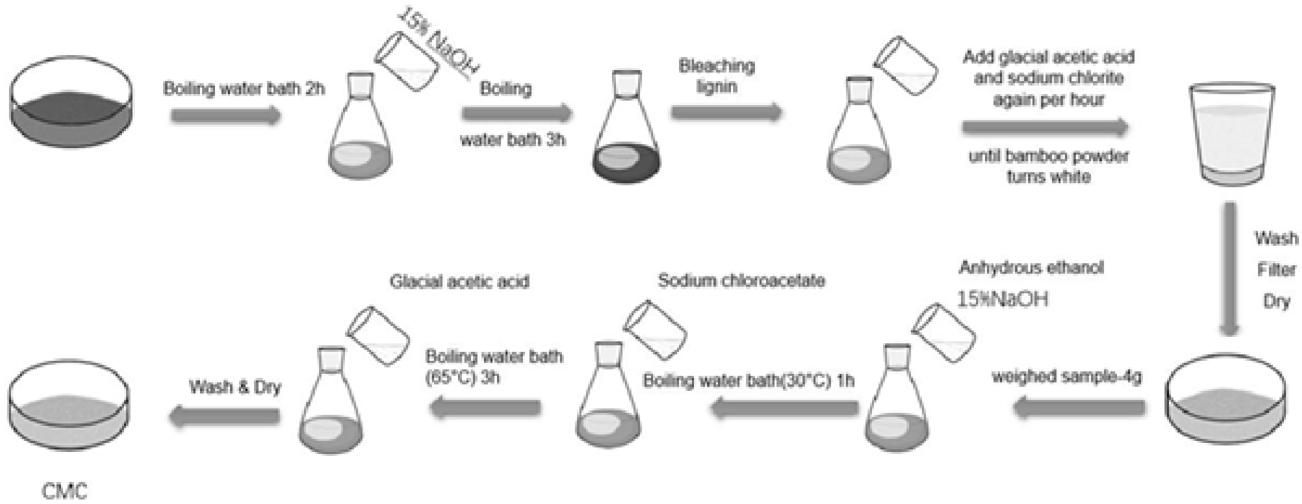

Figure 1: Preparation flow chart for CMC.

Extraction and dissolution of jute cellulose: The research team of Xu [5] used jute fiber as the basic raw material for cellulose-forming film. Preparation of cellulose solution, according to sodium hydroxide, thiourea, urea, water and other methods to prepare cellulose solvent, the jute fiber was subjected to alkali-oxygen pre-defoaming pretreatment and left to stand in the refrigerator. When it is observed that the cellulose solvent has obvious needles precipitated from the water, the prepared cellulose solution can be obtained by adding the pre-defoamed jute fibers with alkali oxygen into the prepared cellulose solvent.

Extraction and dissolution of salix cellulose: Niu [6] pulverized and dried salix, added wood powder into nitroethanol mixture, and placed it in a boiling water bath to react to obtain salix cellulose, which was dissolved by ionic liquid (BMIM)Cl.

Extraction and dissolution of halocellulose from bagasse: After the research of Yang [7], the preparation and extraction method of bagasse holocellulose is to add bagasse solution into sodium chlorate solution, and then extract the bagasse holocellulose obtained by heating, filtering, washing, drying and other steps. After preparation, extract the zinc chloride bagasse solution with a mass fraction of 70%, add a certain amount of holocellulose in it, place it in a water bath, heat and stir until it dissolves for a certain period of time to obtain a holocellulose solution.

Laboratory preparation method of cellulose membrane

The cellulose film is prepared in the laboratory mainly by dissolving the cellulose and spreading it on the mold by the casting method, and then immersing it in the corresponding coagulation liquid to finally obtain the cellulose film.

The process of preparing cellulose film from bamboo harvesting residues: A solution (suspension) of sodium carboxymethyl cellulose was prepared with distilled water, placed in a water bath and heated with constant stirring. When sodium carboxymethyl cellulose is completely dissolved in water, add a thickener and continue to stir until it is evenly mixed, then cast on a polytetrafluoroethylene mold to form a film.

Preparation method of jute cellulose film: The defoamed cellulose solution was poured onto a glass plate, and a film was formed by scraping evenly with a glass rod, and then allowed to stand at room temperature for 20 minutes. Then, a sulfuric acid solution with a mass fraction of 5% is selected as the coagulation liquid, and the coated glass plate is immersed in the solution at room temperature. After the film is formed, the cellulose film is taken out from the coagulation liquid, rinsed with deionized water, and naturally dried at room temperature for use.

Preparation method of salix cellulose film: Take [BMIM] Cl and heat it to a molten state, and after it is completely melted, add it to salix cellulose, and stir to fully dissolve the cellulose. The completely dissolved liquid was allowed to stand for 24 hours to fully deaerate, and then it was uniformly cast on a glass plate to form a film, which was immediately immersed in deionized water. Finally, freeze-dry the cellulose membrane to obtain a dry cellulose membrane.

Preparation method of bagasse holocellulose film: Pour the holocellulose solution on a glass plate, scrape it flat with a glass rod wrapped with copper wires of a certain thickness at both ends, and let it stand. The zinc chloride solution of cellulose will form a cellulose gel after being placed. Then soak it in distilled water to obtain a bagasse helocellulose wet film; fix the wet film on a glass plate and dry it naturally at room temperature to obtain a bagasse helocellulose dry film. The dry film has a dense structure and good Mechanical properties, can be used in the field of packaging (Figure 2).

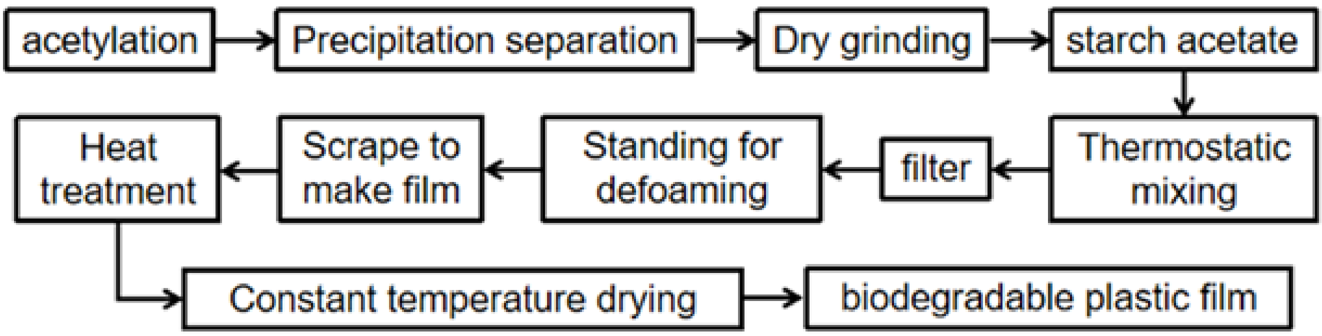

Figure 2: Preparation process of biodegradable plastic film.

Preparation of cornstarch-based cellulose film: After the corn is pulverized, corn starch is obtained for acetic acid acetylation. The light brown transparent acetate solution is directly poured into distilled water, the white precipitate is washed to neutrality, filtered, dried and ground to obtain acetate cornstarch. The main preparation process conditions and experimental operation flow of acetate corn cellulose are consistent with the preparation method of acetate starch. The starch acetate and cellulose acetate obtained by the above reaction are mixed and placed in an iodine measuring bottle respectively, and acetone is added successively as a solvent, tributyl citrate as a crosslinking agent, and dibutyltin dilaurate as an initiator, sodium ethoxide as an accelerator. Then, stir and stand still until completely dissolved and defoamed, and evenly scrape the obtained solution on a layer of flat glass to make a film. Then immerse it in a small amount of ethanol solution and continue to soak for 12 hours and take out the glass plate. Then, it is washed with a small amount of distilled water, and the cellulose/starch-based degradable plastic film can be obtained immediately.

Summary

In the preparation process of cellulose film formation in the above five different plants, except cellulose itself is an environmentally friendly material, almost all additives are used to assist film formation [8]. In the natural degradation process of cellulose membrane, these chemicals may still have certain effects on soil, groundwater system and air. Among them, the preparation method of ionic liquid-dissolved cellulose selected by Niu's team is more green and environmentally friendly than other preparation methods in this paper, which can also become a key development direction in future research. Among them, ionic liquid is a new type of green solvent, which has strong dissolving ability to cellulose, high stability, and no pollution to the environment [9,10]. In addition, the selection of raw materials for cellulose extraction should avoid using crops as the main raw material. Zhang's team [11] used corn starch as the raw material, although the material itself is an environmentally friendly material, but today's hunger problem is still an international problem, so choose non-crop crops. Plants have broader development prospects as raw materials for cellulose extraction and are more in line with the current world development trend.

Performance Analysis of Cellulose Film

Since the research direction of this paper is the application of cellulose film in wet garbage bags, the mechanical properties of the film are one of the most important indicators. At the same time, the biodegradability of cellulose membrane is also an important reference index.

Analysis of mechanical properties of membranes

The tensile properties of sodium carboxymethyl cellulose degradable films can be tested in accordance with the national standard "Testing Methods for Tensile Properties of Plastic Films (GB 13022-1991)" [12]. According to the analysis of Li's team, in the process of processing and preparing CMC, due to the residual lignin and other chemical substances in the raw bamboo powder, the solubility of CMC in water is limited, so the concentration and viscosity of the prepared solution are low. It turned out that it was not easy to make a film. Further experiments show that a certain thickener needs to be added to the solution to improve the properties of the sodium carboxymethyl cellulose film [13]. With reference to other research, the team found that the common thickener sodium alginate is a natural polymer composed of alternating D-mannuronic and L-guluronic segments, and has good biodegradability, biochemical compatibility and good film-forming properties, are widely used as food chemical additives, biomedicine and other materials [14]. Among the natural polymer materials with degradable raw materials, starch is a wide-ranging choice, and the mechanical properties of the films obtained by mixing it with CMC are improved. After analyzing the mechanical properties of the jute cellulose film, it was found that the effect of the jute fiber mass fraction on the breaking strength and elongation at break of the cellulose film is shown in Figure 3. It can be seen from (Figure 3) that with the increase of the mass fraction of jute fiber, the breaking strength and elongation at break of the jute cellulose film first increased and then decreased. After calculating the crystallinity of jute cellulose film, it is found that with the increase of fiber mass fraction, the crystallinity of the film itself increases. When the fiber mass fraction is 5%, the film crystallinity reaches the maximum value; increased, the crystallinity of the film showed a downward trend. This is because the viscosity of the cellulose solution is proportional to the fiber mass fraction, and the enhanced interaction of fiber macromolecules makes it easier to form intermolecular hydrogen bonds, so that the initial semi-rigid structure of intermolecular is converted into a tightly ordered arrangement. The higher the degree of molecular chain order, the more crystals the cellulose film produces during solidification, so the crystallinity and breaking strength of the jute cellulose film are proportional to the fiber mass fraction.

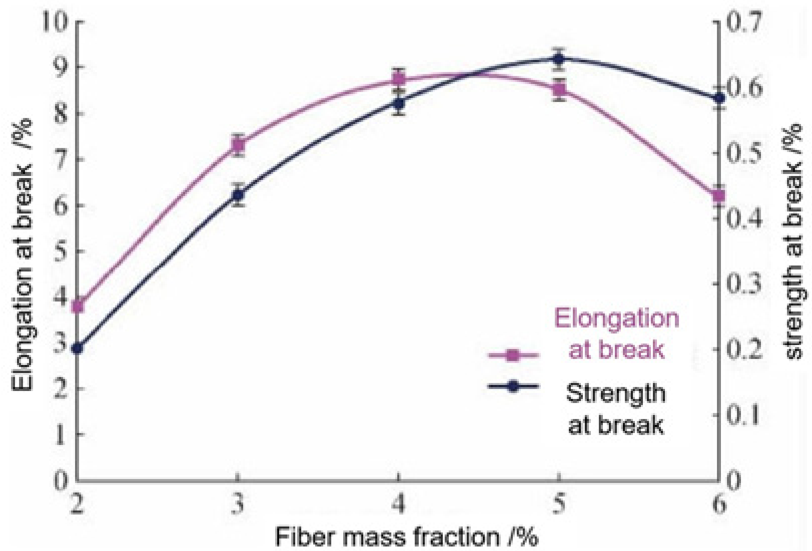

Figure 3: Effect of fiber mass fraction on mechanical properties of jute cellulose membrane.

The static mechanical tensile analysis of Salix cellulose film showed that the maximum tensile stress value increased with the increase of cellulose content (Figure 4). Since cellulose is primarily responsible for tensile strength, the greater the tensile stress that a cellulose film can withstand is proportional to the cellulose content, and the crystallinity of the film and its associated tensile properties also increase with the cellulose content and strengthen. It can be proved from (Figure 5) that the strength of the cellulose film will also increase when the binding between celluloses is tighter. The analysis of mechanical properties of bagasse cellulose membrane is special. Since the dry film obtained from bagasse cellulose film is relatively brittle, it will lack toughness without plasticization, and has little use value. Therefore, it is necessary to soak the film in glycerin and then balance it in a constant temperature and humidity box, and then measure its mechanical properties. performance. After testing and analysis, it can be found that the dry film with 5% cellulose has the highest tensile strength after being soaked and plasticized in glycerol (Figure 6). After analyzing the mechanical properties of the starch-based film, it was found that the tensile stress and elastic modulus of the film decreased with the increase of the starch acetate content, and the elongation at break increased and then decreased. When the ratio of starch acetate to cellulose acetate is the same, the mechanical index of the plastic film is good, but the elongation at break of the film in glycerol is not perfect. According to Zhang's team [15], this is due to the inherent factors of starch cellulose-based materials that determine this defect. In the follow-up research, we try to improve this performance by mixing different polymer materials.

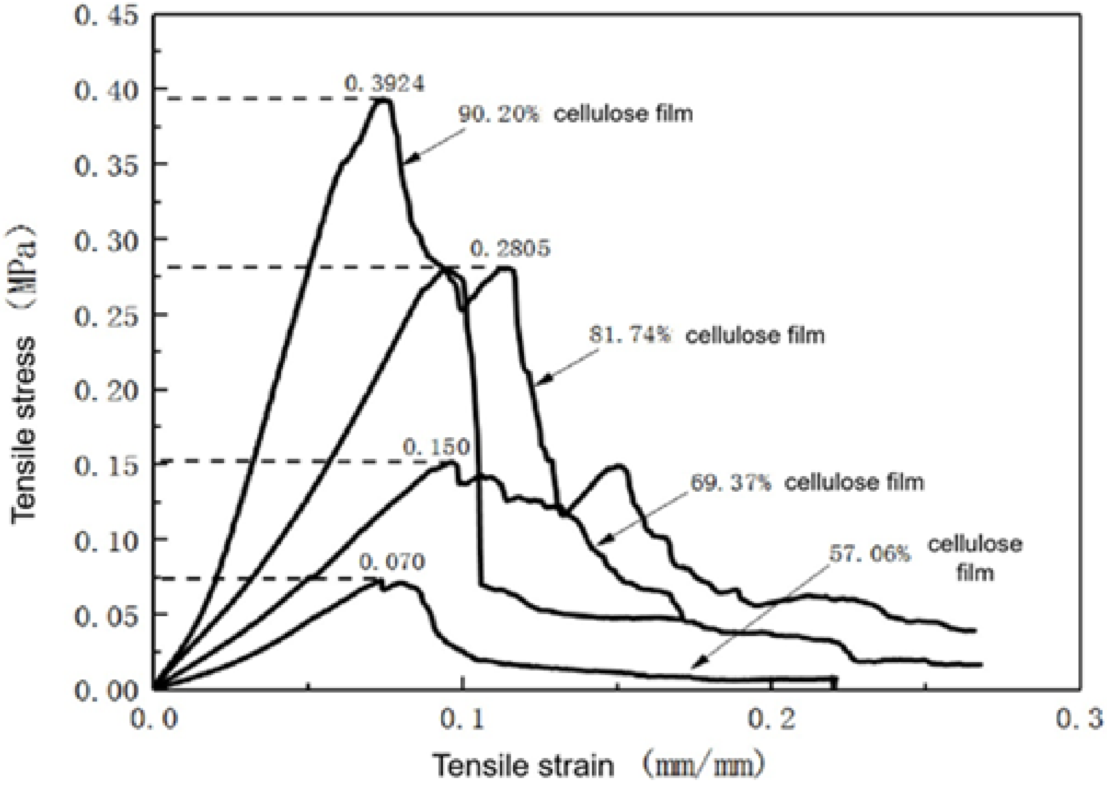

Figure 4: Mechanical properties of different cellulose content films.

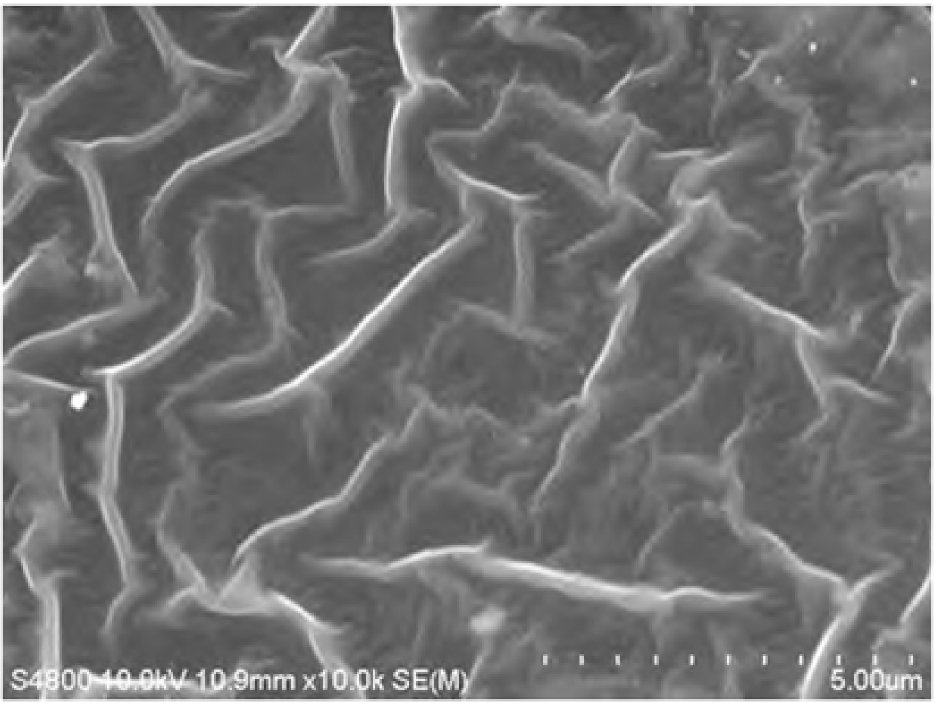

Figure 5: SEM of cellulose membrane formed by one time treatment.

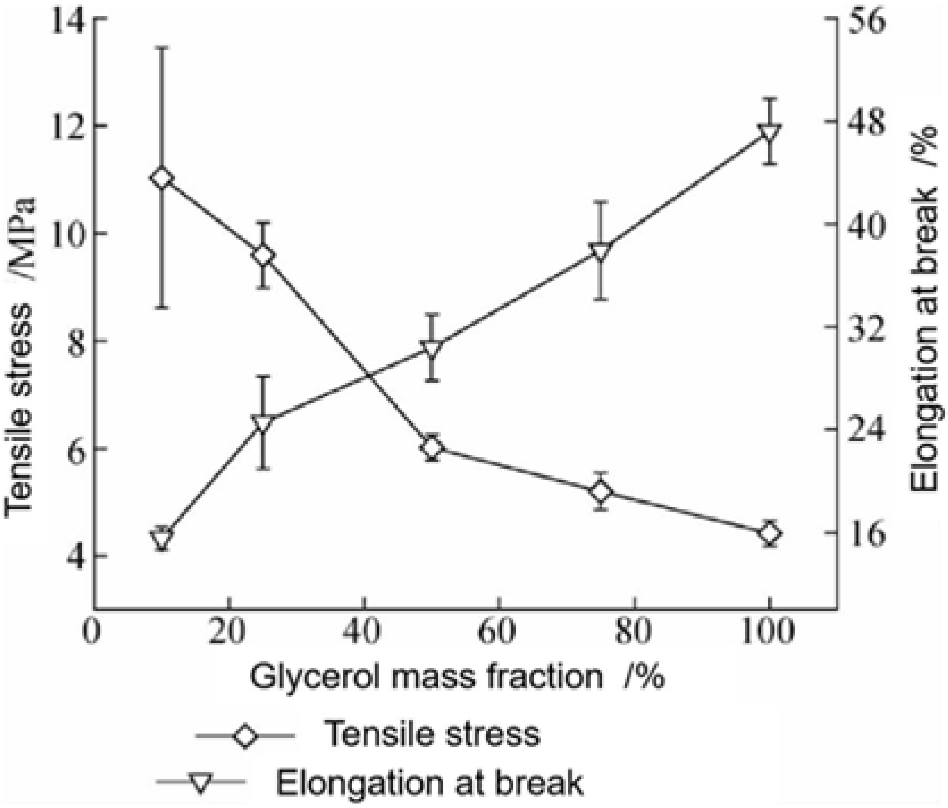

Figure 6: Effect of mass fraction of glycerin on mechanical properties of film.

Degradability of cellulose film

The current biodegradability test can refer to ISO 16929:2002 (Determination of the degree of disintegration of plastic materials under the defined composting pilot conditions), and the collected test data of the relevant cellulose film biodegradability are as follows: Regarding the biodegradability of jute cellulose membrane, the decomposition pressure of microorganisms in the soil increases with the increase of the mass fraction of jute fibers in the cellulose membrane, so the degradation rate of the cellulose membrane gradually decreases. The soil filling degradation behavior of cellulose membranes with different components is roughly the same, showing that the initial degradation rate is low. The contact surface area is increased, so the degradation rate is increased to a certain extent [16]. After analyzing the results of the biodegradation experiment of starch-based soil burial, the experimental results of the biodegradation rate of the biodegradable plastic film of each component in 120 days are shown in Figure 7 [17]. It can be seen from (Figure 7) that the biodegradation rate of the biodegradable plastic films of different components is the fastest in the first 30 days, and the degradation rate of the degradable plastic films of each component gradually becomes flat after 60 days. The degradation rate did not change significantly during the day. At this time, due to the hydrolysis of the membrane and the action of microorganisms in the soil, the membrane had already presented a loose and broken form, and the biodegraded polymer contained a large amount of cellulose and other starch substances. In theory, it will gradually decompose under the combined action of a small amount of water and microorganisms within a few months [18].

In Figure 7, the application of acetate starch in the component can effectively improve the soil burial biodegradation rate of the film, and with the further increase of acetate starch content, the soil burial biodegradation of the plastic film in the same time period. The rate is also increased, which theoretically indicates that the biodegradable film added with acetate starch material has better soil burial biodegradability [19]. According to the comparative analysis of mechanical properties, the content of cellulose is proportional to the strength of the film, but when the content of cellulose is too high, the breaking strength of the film will decrease. At the same time, the biodegradability of the membrane also decreased with increasing cellulose content [20,21].

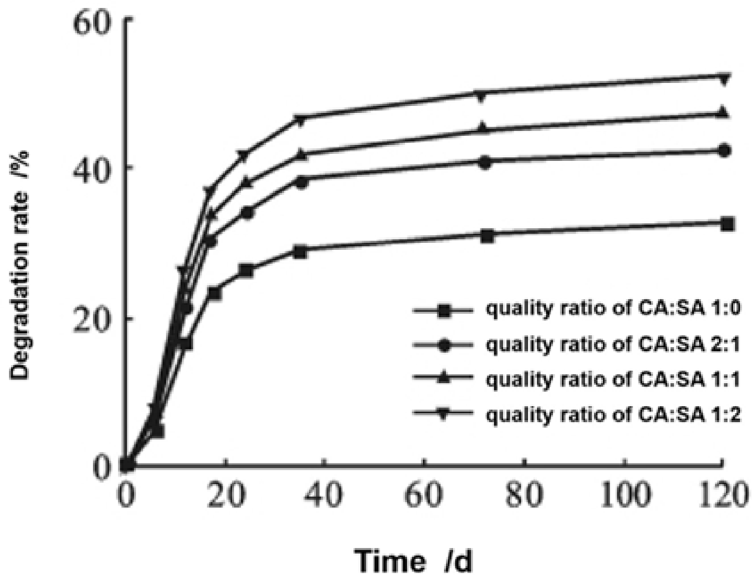

Figure 7: Soil-burial degradation rate of biodegradable plastic films in different composition.

Conclusion

Experiments can prove that the cellulose in plants can be extracted by drying and grinding, and the cellulose solution can be obtained by dissolving it in an alkaline solvent or ionic liquid. operability. At the same time, it also has certain mechanical properties, and the biodegradability of starch-based cellulose film and jute cellulose film is proved by the degradability of soil landfill. The biodegradability of cellulose membranes from bamboo harvesting residues cannot be demonstrated due to lack of data. At present, the research content and themes of cellulose films range from single-material film formation, mechanical property analysis, and step development to the preparation of multi-functional composite films and their application activity analysis of preservation, degradation, and bacteriostasis. The research frontier fields of cellulose-based food packaging films mainly focus on food packaging technology, material science, food safety, etc., especially hot spots such as antibacterial films, nanocellulose, chitosan, composite films, active packaging, and mechanical properties.

Sirviö et al. [22] used a solvent casting method to prepare a cationic wood fiber film with high oxygen barrier properties, light transmittance and good mechanical strength, which could inhibit Staphylococcus aureus and Escherichia coli. Bansal et al. [23] modified the nanocellulose fibers extracted from bagasse and blended them with chitosan to prepare a composite membrane with good antibacterial properties. Hai et al. [24] used bamboo nanofibers and chitin to prepare a new type of nanocomposite with good thermal stability, suitable tensile strength, and complete biodegradation. Feng et al. [25] used bacterial cellulose to prepare a composite membrane that had a significant inhibitory effect on Listeria monocytogenes. Wang Tingting et al. [26] loaded rhodopsin on bacterial cellulose/chitosan composite materials to prepare a nanocellulose film with antibacterial function. It can be seen that fiber-based composite materials have been widely used in food preservation and antibacterial packaging materials, active packaging materials and high barrier packaging materials.

Conjectures and Prospects

Combining the above research and conclusions, the following three conjectures are made: First, the main cellulose is currently extracted from plants. In the follow-up research, we can try to add related materials to improve the waterproofness of the membrane to cope with the high moisture content of wet garbage and high-water content. The status quo that the composite paper bag is easily soaked. Secondly, whether pre-mixing and crushing can be used in the process of landfill degradation of the film to accelerate the decomposition of the cellulose film. Thirdly, in the process of landfill degradation, the decomposition of biodegradable bags will release substances such as carbon dioxide and water, resulting in soil acidification, so whether the alkaline cellulose solvent used in film making can be used as lye to pre-spray the soil before landfilling. In order to improve the degradation rate and alleviate the problem of soil acidification. In the future, the green and environmental protection of cellulose solvents needs to be studied, and a solvent that is simple and easy to operate and has good dissolution effect is developed, so that it can be industrially produced. At the same time, the state should take certain measures to support and encourage the research and development of sustainable polymer materials, so that biodegradable materials and products can enter the market as soon as possible.

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