International Journal of Scientific Research and Engineering Development-– Volume 8 Issue 1, Jan-Feb 2025
Available at www.ijsred.com

RESEARCH ARTICLE OPEN ACCESS

Strengthening Biomass Waste-Derived Bioplastics with the Addition of Cellulose Nanofibers

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Abstract:

The widespread use of conventional plastic packaging has led to significant environmental challenges, including waste mismanagement, plastic accumulation, and environmental degradation. Biodegradable polymers, particularly starch-based films, offer a promising alternative to replace conventional plastics like HDPE due to their comparable mechanical properties. However, extracting starch from food sources raises concerns about exacerbating global food waste, creating a need for more sustainable approaches. Waste biomass, such as banana peels and sugarcane bagasse, presents a viable solution, providing renewable sources of starch while addressing food waste issues. Additionally, the incorporation of cellulose nanofibers (CNF), also derived from waste biomass, significantly enhances the mechanical properties of starch-based films, improving their suitability for packaging applications. This study conducts a comprehensive literature review, analysing three biodegradable film formulations produced from waste biomass and comparing their mechanical properties to identify the most effective and sustainable alternative to plastic packaging.

Keywords — Biodegradable, CNF, Extraction, Mechanical properties, Starch, Waste

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I. INTRODUCTION

Non-biodegradable materials for packaging purposes have raised the number of environmental impacts and concerns [1]. Large amounts of glass, plastics, paperboard, steel, and aluminium are used globally to manufacture food packaging [2]. These materials are unsustainable as they cannot be maintained in the environmental cycle while also causing harm to the environment because they cannot be broken into smaller particles by natural processes [3]. For example, plastic production has doubled over the last two decades globally, reaching approximately 460 million metric tonnes per year [4]. As a result, plastic waste in the environment is proliferating, with 40% of the world’s waste ending up in open dumpsites. [5]. Furthermore, packaging industries have become one of the largest primary sources of plastic waste accumulation [6]. In India, packaging constitutes 59% of total plastic consumption, while in Europe it is 40% [7]. Additionally, 90% of plastic solid waste is mismanaged and dumped openly in India [5], which leads to environmental decay and microplastic aggregation in soils.

There is an urgent need for sustainable packaging materials at an equivalent cost of plastic packaging to protect the environment from degradation [8]. Biodegradable materials can come from multiple natural sources of food-based, plant-based and microbial polymers [6, Fig. 1]. However, some food-based sources are not as appropriate and sustainable. One third of the food produced globally for human consumption is wasted [9]. And if used to produce polymers, not to solve issues such as the global hunger crisis [10], the process of producing these ‘sustainable’ polymers is not benefitting the environment. Therefore, food and biomass waste are suitable alternatives as the waste is getting recycled in order to create the biodegradable polymers. There are some solutions for materials that focus on replacing conventional packaging methods such as seaweed, coconut husk, however, among these biodegradable materials, starch is renewable, inexpensive, abundant, and mechanically strong to act as an alternative for conventional packaging [6]. Consequently, non-biodegradable plastics are being slowly replaced with these natural biopolymers such as PLA (polylactic acid) and starch-based plastics [11].

Starch is extracted from food-based sources majorly, but to make the final polymers and nanocomposites sustainable, starch is also extracted from food waste materials. By reusing and recycling the waste, the carbon footprint is reduced drastically as well. This further reduces the impact towards the environment.

Moreover, to successfully replace plastic packaging with biodegradable starch polymers for food packaging, certain mechanical properties must be matched. High-density polyethylene (HDPE), a plastic used widely for packaging purposes, has a high tensile strength value, while also being flexible and durable [12]. Due to its high elongation at break, it can handle impacts and stress, making it a suitable packaging material. However, the goal is to replace these materials. Starch polymers often have high ductility and are non-toxic, odourless and semipermeable to moisture [13]. However, they have limited strength and a high absorption rate of water. Modification and reinforcement increase the thermal and mechanical properties of the starch film, a process which has become widely researched [6]. This can be done by chemical and enzymatic methods which increases the starch's functionality as a packaging material.

Recently, cellulose nanofibers (CNF) have become a common filler to modify starch-based polymers to produce nanocomposites. Cellulose is a largely abundant polymer and has several applications due to its biodegradability and biocompatibility with other polymers [6]. The incorporation of CNF in starch polymers has shown improvements in largely mechanical properties like tensile strength and elongation at break [14]. These values increase with the addition of CNF fillers and its permeability and water moisture content lower as well. Cellulose nanofibers are extremely low density and light weight in comparison to other artificial fillers, and they are easily extracted from waste biomass – consuming less energy for extraction.

Sinha et al., (2021) [8], states that "India wastes as much food as the whole of United Kingdom consumes". This waste is composed of 32% carbohydrates which includes starch and CNF [8, Fig. 1]. The source of this waste often consists of fruit and vegetable waste that is openly dumped. In India, banana and sugarcane are majorly consumed due to cultural and agricultural reasons. For example, India is the world's largest banana producer and due to its consumption, approximately 3.5 million tonnes of banana peel waste is produced each year [15]. 30-40% of the total weight of each banana is its peel [12], and this can be used to extract starch and cellulose. The peels have around 9% concentration of cellulose especially. Sugarcane is also one of the main crops cultivated in India. 90 million tonnes of sugarcane bagasse (waste residue) is produced annually in India [16]. This bagasse has 32-45% of cellulose content that can be extracted to be used to produce nanocomposites [13]. Therefore, both banana peel and sugarcane bagasse are suitable waste products that can be used as sources of starch and CNF.

As a result, this research paper discusses the potential of using starch and cellulose extracted from biomass waste, specifically banana peel and sugarcane bagasse, for the development of biodegradable films. The study begins with the exploration of the fundamental characteristics of starch and cellulose, followed by a review of existing research on extraction methods and their feasibility from waste biomass. Next, a comparative analysis of different formulas is put together to discuss and evaluate the mechanical properties. Lastly, the paper assesses the sustainability of these films by focusing on the impacts of the extraction processes. The structured approach helps to identity packaging as a suitable application for these films.

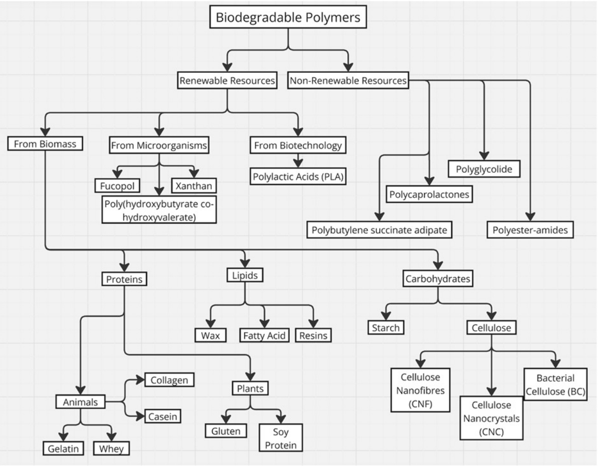

Fig. 1 A flow chart showing types of biodegradable polymers [Adapted from Bangar et al, 2021 (6)]

II. LITERATURE REVIEW

A. *Starch Extraction*

Starch granules consist majorly of two polysaccharides – amylose and amylopectin. Amylose has linear chains of α-(1,4)-linked d-glucose residues, which are interconnected through amylopectin's α-(1,6)-glucosidic linkages, thus forming branches in the polymers [17]. Starch granules are semi-crystalline as they contain both crystalline and amorphous parts [9]. The combination of linear and branched structural features of starch make it a unique source of energy storage in plants and creates applications for the extraction of starch in different fields.

Starch is a biomass that can get easily extracted due to its structural properties. The biotechnology and methods of starch extraction are improving to its best quality annually. There are multiple methods that can be used to extract starch from banana peels since banana peels have a high concentration of starch. Isolation of starch through heat moisture treatment, wet extraction method, and ultrasound assisted enzymatic extraction are some commonly used methods.

Wet extraction method is the isolation of starch through the preparation of banana peel powder firstly [18]. The powder is first mixed with water in a 1:5 ratio and soaked at 30°C for three hours. The mixture is then blended and passed through a sieve to separate the filtrate and the solid residue. The residue was further treated with ethanol (70%) and then with 0.1 M NaOH to remove impurities, leaving behind solid material that was set aside for nanocellulose extraction. The collected filtrates were centrifuged, and the supernatant was removed. The remaining starch solution was then washed with sodium hydroxide and deionized water, filtered, and freeze-dried, with the final dried starch stored at -5°C for later analysis.

Ultrasound-assisted enzymatic extraction (UAEE) [19] involves the use of enzymes and ultrasound waves to enhance the extraction of starch from banana peel waste. Initially, enzymes such as amylase or cellulase are applied to break down the plant cell walls, which releases the starch granules. Simultaneously, ultrasonic waves generate cavitation, where the rapid formation and collapse of microbubbles create localized shear forces that help disrupt the cell structure. These forces enhance the penetration of enzymes into the material and facilitate the release of starch granules from the banana peel matrix. Following the treatment, the starch is separated by filtration and centrifugation before being dried for further use. The films can be analysed using X-ray diffraction analysis to check its crystallinity.

TABLE I
METHODS OF STARCH EXTRACTION [15, 16]

Method	Basic Working Principles	Advantages	Disadvantages
Wet extraction method	- Banana peel powder is mixed with water and filtered to separate residue. - Filter is treated with ethanol and NaOH to remove impurities, and then centrifuged to isolate starch. - Starch solution is then washed, and free-dried to store. [15]	- Small-scale application & cost effective. - High yield and purity of starch obtained. - 35^0C low temperature used.	- Large amounts of water and energy needed. - Wastewater is created that needs proper disposal.
Ultrasound assisted enzymatic extraction	- Certain enzymes are applied to break down plant cell walls of the banana peel, releasing starch granules. - Ultrasound waves help disrupt cell structure, allowing for more enzyme penetration. - Centrifugation and filtration help isolate	- Enzymes and ultrasound enhance starch yield. - Does not require chemicals for purification. - 35^0C low	- Specialized equipment is required which is expensive. - Incorrect application of equipment can reduce yield.

	starch. [16]	temperature used.	

B. Cellulose Extraction

Cellulose is the primary structural component of the cell wall in lignocellulosic plants [20]. It is a naturally occurring polysaccharide which consists of glucose units bounded by β-1,4-linkages. These occur in linear chains, fabricated into microfibrils which are stabilized by hydrogen bonds between hydroxyl groups along the chain. This structure with the hydrogen bonds gives cellulose its mechanical and thermal stability.

CNF is produced by the breaking down of cellulose into a nanoscale, with a small diameter of 5-50 nm. CNF is extremely lightweight and strong due to its aspect ratio. This is also because of its structural composition of the hydrogen bonds. Therefore, CNF is a suitable filler for starch to increase its mechanical properties to create sustainable packaging. Recently, researchers have found several methods to extract CNF from cellulose in a way to obtain high purity such that its properties remain the same. This can be done from sources of banana peels and sugarcane bagasse since the concentration of CNF is highest in these two.

CNF can be extracted from sugarcane bagasse (SCB) using the alkaline treatment, mild acid hydrolysis, ultrasonication methods [21]. 2% NaOH was added to the SCB after which the mixture was heated in a water batch to 80 °C for 5 hours to filter out the non-cellulosic substances in the bagasse. This was repeated with 12% NaOH at 80 °C for an hour more. Next, the solution is added in aqueous hydrogen peroxide and filtered to remove impurities. After the process was repeated several times, it was stored in a refrigerator at 4°C. the cellulose nano fibres were now subjected to ultrasonication for 2 hours, followed by centrifugation for 15 minutes to separate the nanofibers. This helped produce the CNF in the correct quantity.

Sodium Carbonate (Na_2CO_3) treatment allows for the extraction of CNF from sugarcane bagasse as well [22]. After treating the SCB powdered solution with Na_2CO_3 at elevated temperatures, the non-cellulosic substances like lignin are removed. After this initial extraction, researchers use an ionic liquid called 1-n-butyl-3-methylimidazolium chloride to further process the fibres into CNF. The diameter of CNF is therefore around 4-35 nm. This is an environmentally friendly way to produce CNF in high concentrations.

CNF is also extracted from banana peels. Similar to the extraction of cellulose from SCB using the alkaline treatment and mild acid hydrolysis, after the hydrogen peroxide is used to remove impurities, sulfuric acid of different concentrations is added to break down the cellulose into nanoscale fibres [23]. After which, high-pressure homogenization is used to reduce the diameter and size of the fibres to produce CNF. This allows for the application of CNF in packaging.

Enzymatic treatment to extract CNF from banana peels is another effective method since it obtains a high yield. Xylanase is an enzyme that can remove the non-cellulosic components like hemicellulose. Enzymatic hydrolysis targets the breakdown of these substances to isolate the cellulose nanofibers from the cellulose present in the banana peels. Usually, an enzyme concentration of 70 U/g of banana peel, a substrate concentration of 15%, a pH of 6.0, and a temperature range of 35–55°C have been shown to favour enzymatic hydrolysis [24].

TABLE II
METHODS OF CELLULOSE EXTRACTION [18, 19, 20,21]

Source	**Method**	**Basic Working Principles**	**Advantages**	**Disadvantages**
Sugarcane Bagasse	Alkaline treatment, mild acid hydrolysis, ultrasonication [18]	- SCB treated with 2% NaOH at 80°C for 5 hours, then 12% NaOH at 80°C for an hour. - Hydrogen peroxide treatment and filtration to remove impurities - Ultrasonicated and centrifuged to isolate CNFs.	- Less chemical waste as low concentrations of NaOH. - Ultrasonication improves yield of CNF. - High-quality CNF produced.	- Time consuming as many steps involved. - Ultrasonication needs high energy. - Could impact environment due to chemicals used.

	Sodium Carbonate (Na_2CO_3) treatment [19]	- Treated with sodium carbonate to remove impurities. Processed with ionic liquid to produce fine CNFs.	- Environmentally friendly. - Produces CNF with a small diameter. - Reduces need for harsh chemicals.	- Ionic liquid treatment could be expensive and is specialized. - Time consuming due to multiple steps.
Banana Peel	Chemical and Mechanical treatment [20]	- Hydrogen peroxide is used to remove impurities. - Sulfuric acid breaks down the fibres. - High-pressure homogenization is used to reduce size of CNFs.	- High-pressure homogenization is an extremely effective method for the production and extraction of CNFs.	- High-pressure homogenization is energy-intensive and is specialized. - Time consuming.
	Enzymatic Treatment [21]	- Xylanase enzyme is used to break down non-cellulosic substances. - Enzymatic hydrolysis requires specified concentrations.	- High yield of CNF from enzymes. - Sustainable method as it uses enzymes instead of chemicals. - Allows for fine control over method.	- Requires precise and specialized training for the parameters involved. - Specific enzymes may be expensive

III. METHODOLOGY

The literature review methodology follows a structured approach to gathering and analysing relevant research related to the central topic on biodegradable packaging. A thorough search of scholarly articles is conducted using Google Scholar, focusing on terms like bioplastics, starch, biomass waste, and CNF. The process involves summarizing key points from selected sources, paraphrasing findings, and integrating them to provide a comprehensive understanding. By evaluating and synthesizing the literature, the review highlights knowledge gaps and suggest potential solutions, ensuring proper citations to avoid plagiarism.

Building on this methodology, the review narrows its scope to biodegradable packaging derived from waste biomass. A focus is placed on banana peels and sugarcane due to its abundance as a product in India as well as the large amounts of waste it creates. This selection was also guided by their high starch and cellulose content thus they serve as a potential material for packaging. Studies on the mechanical properties of starch films reinforced with CNF were examined while also integrating environmental impacts and considerations. Compared to research papers by Bangar et al., (2021) [8], and Pelissari et al., (2017) [25], this paper works on using two different sources of cellulose while also presenting a comprehensive sustainability analysis on the extraction methods and their impacts on the environment.

IV. RESULTS & DISCUSSION

A. Comparison of Different Starch and Cellulose Mixture formula in regards of Mechanical Properties

TABLE III
MECHANICAL PROPERTIES [24, 25, 26, 27,28]

Properties	**Plastics - HDPE**	**Formula 1 [25]**		**Formula 2 [26]**		**Formula 3 [27]**	
		Starch	**CNF**	**Starch**	**CNF**	**Starch**	**CNF**
Composition	-	95%	5%	92 %	8%	90 %	10 %
Source	-	Banana	Banana Peel	Tapioca	Sugarcane Bagasse	Cassava	Wheat Straw
Purity	-	94.8%	-	98 %	-	-	-
Width of CNF	-	10.9 nm		23.2 nm		34.0 nm	
Extraction Method of CNF	-	Chemical and Mechanical Treatment		Alkaline treatment, mild acid hydrolysis, ultrasonication		Alkaline treatment, mild acid hydrolysis, ultrasonication	
Tensile Stren	7.6 – 43.0 MPa	8.90 MPa		3.50 MPa		11.62 MPa	

gth	[28]			
Elongation at Break	3.2 – 2230.0% [25]	25.90%	16.00%	4.66%
Young's Modulus	600 – 1500 MPa [29]	768.60 MPa	72.00 MPa	490.16 MPa

The starch film filled with CNF can be compared to HDPE's mechanical properties to make sure that conventional packaging can be replaced by a more sustainable one. Tensile strength, elongation at break, and Young's modulus are three key parameters for the comprehensive understanding of the mechanical properties of different materials under stress and strain. These helps demonstrate whether the material can be used to develop packaging films and be handled under storage and processing conditions [30]. Several studies have been conducted to study the effects of cellulose enforced starch films as packaging materials and its mechanical properties.

Formula 1 (F1), formula 2 (F2), and formula 3 (F3) are taken from different studies conducted that are based on the mechanical properties of CNF enforced starch films. They are compared to HDPE's properties in order to find the best combination of biomass waste that can be the source of starch and cellulose to create sustainable packaging.

a. Effect of CNF Width on Mechanical Properties

Different methods of extraction of CNF lead to different yields and widths of the cellulose fibres. According to Lim et al., (2012) [31], the width of the CNF affects the mechanical and physical properties of the material as the aspect ratios, surface areas, and dispersion of the structure change according to the width and diameter of the CNF.

Referring to *Table 3,* F1 has a CNF width of 10.9 nm while having a tensile strength of 8.9 MPa. F2 which has a CNF width 23.2 nm has 3.5 MPa tensile strength. On the other hand, F3 has the biggest CNF width of 34 nm and also the highest tensile strength of 11.62 MPa. A bigger width of CNF in a mixture with starch could have higher tensile strength properties in comparison to other widths as it will have a higher aspect ratio which means that the film has a stronger network of entangled fibres which can contribute to increased load efficiency as well [28].

Elongation at break is a measure of a material's ability to stretch or deform before breaking, expressed as a percentage of its original length under tensile stress [32]. Despite F1's width being the smallest at 10.9 nm, it has the highest elongation at break of 25.9% compared to F2 and F3's 16% and 4.66% respectively. Researchers say that elongation at break could be higher for smaller widths of the CNF as the area-to-volume ratio is better which leads to the interaction and dispersion of the particles to be improved [29]. The stretch distribution of the particles therefore is more even with smaller sized CNF, which allows the film to experience higher stress before breaking.

Furthermore, Young's modulus is another parameter of mechanical properties that gives a holistic understanding of the film's strength. It is the ratio of stress to strain and measures the material's stiffness. In *Table 3,* F1 has the highest Young's modulus of 768.6 MPa while F2 has only 72 MPa. F1 has the smallest width of CNF and therefore its particle attraction and dispersion allow it to excel in this parameter. Width does affect the value of the Young's modulus as the particles of the starch and CNF interact at different levels if the aspect ratios vary [33]. Overall, F1 has the highest mechanical properties despite its width being the smallest. This varies which means that the width does affect the mechanical properties of a material as it plays with the aspect ratios, interaction and dispersion of the particles, causing different bonds as well.

b. Effect of Extraction Method on CNF Yield Width

Referring to Table 2 the extraction methods of CNF, there are multiple methods that yield different widths of the fibres. Extraction methods vary because they use different substances and treatments to extract the nanocellulose. F1 is yielded from chemical and mechanical treatment that uses hydrogen peroxide, sulfuric acid and high-pressure homogenization (mechanical method) [20]. This method gives a CNF width of 10.9 nm. However, F2 and F3 use alkaline treatment, mild acid hydrolysis, and ultrasonication to yield larger widths of CNF. This includes 23.2 nm and 34 nm respectively.

Chemical and Mechanical treatment yield smaller CNF as it intensively breaks down the cellulose and removes the non-cellulosic materials. The chemical hydrolysis using hydrogen peroxide and sulfuric acid targets majorly on the amorphous parts of the nanocellulose which creates a way for better separation of the nanofibers by reducing its bonding and improving the dispersion [34]. The range of separation can be as high as around 80-90%, which explains why a smaller width of CNF is extracted [37]. The high-pressure homogenization allows for the further fragmentation of the nanofibers by the application of shear forces and pressure [20].

However, the alkaline treatment, mild acid hydrolysis, and ultrasonication yield larger widths of CNF as this method focuses on the removal of hemicellulose more than the shear breakage of the fibres. NaOH and mild concentrated acid is used instead of a highly concentrated sulfuric acid to remove these impurities. Ultrasonication, although reduces fibre agglomeration efficiently, is less precise in separating the cellulose into nanoscale dimensions in comparison to the mechanical method of high-pressure homogenization [31]. As a result, the two extraction methods vary based on the kind of substances used, therefore yielding CNF of different widths.

c. Achieving HDPE Mechanical Properties using Sustainable Formulas

HDPE plastics require a range of values that should be matched when packaging is formed in industries. F1, F2 and F3 have specific mechanical properties that have been tested by different studies. According to Table 3, F1 is the only formula that fits into the range of mechanical properties exhibited by HDPE. These formulas must achieve the HDPE mechanical properties as HDPE is not only one of the widely used materials for packaging applications but also there are technical standards for packaging to be manufactured [35]. Starch and CNF are alternative materials. If combined together in the right ratios, they can act like a more sustainable form of conventional packaging.

If the mechanical properties do not match conventional packaging standards, then they are usually not considered by manufacturers as they may not be flexible or strong enough to experience the stress and pressure that packaging undergoes in multiple processes [34]. As a result, mechanical properties must be considered as important when producing an alternative form of biodegradable polymer films to replace HDPE packaging.

B. Sustainability Analysis

Biodegradability refers to the capacity of a material to be broken down by natural biological processes into simpler elements [36]. Packaging industries require their materials to be biodegradable and sustainable in order to prevent harm towards the environment. Non-biodegradable polymers often leave a long-term impact on the environment; therefore, several studies are being brought upon to investigate the best eco-friendly material for packaging purposes to prevent the degradation of the environment. Modified polymers such as starch are fully biodegradable in comparison with conventional HDPE packaging [37]. The filler of CNF makes the film more sustainable as CNF is more porous which helps in the degradation of the material [38].

Other than the alternative materials of starch and CNF, the fabrication techniques used to extract these substances also plays an important role in determining the level of sustainability of a film. F1 uses chemical and mechanical treatment to extract CNF while F2 and F3 use alkaline treatment, mild acid hydrolysis, and ultrasonication.

Chemical and mechanical treatment uses high concentrations of sulfuric acid and hydrogen peroxide [Table 2]. This process generates a significant amount of chemical waste that requires proper disposal of acid waste [39]. The emissions associated with these processes can lead to harmful impacts on the environment – especially contributing to air pollution. Handling highly concentrated acids also raise several safety concerns. Furthermore, high-pressure homogenization requires extremely large amounts of energy which needs electricity as well [40]. Often, the energy needed is not sustainable enough.

Alkaline treatment and mild acid hydrolysis, although less chemically intensive, relies significantly on large quantities of NaOH and mild acids. The alkaline waste generated from these processes can pose as challenges for disposal and can lead to increased salinity in water bodies if improperly managed [41]. There is high water and energy use in the ultrasonication step as well. This highlights the concern about its carbon footprint as often the use of high amounts of energy is not sustainable for the environment. Both methods of extraction, although obtaining a high yield of CNF, could be considered unsustainable due to the environmental concerns it holds. However, Alkaline treatment, mild acid hydrolysis and ultrasonication is slightly more environmentally friendly than chemical and mechanical treatment due to the repercussions as described. The environmental impacts that occur due to the second method are less severe than the first as the waste produced is lesser and the energy consumed is less significant. To further exemplify the sustainability of extraction methods, more green alternatives for extraction are being researched by many studies all over.

V. CONCLUSION

Starch based polymers modified with CNF are highly sustainable films that can be used for packaging due to its combined mechanical properties. The application of nanocellulose instead of conventional plastic and HDPE allows for the improvement of biodegradable packaging. Specifically, the tensile strength, elongation at break and Young's modulus of the bioplastic film show similar properties to that of the HDPE. However, there is a concern for the environment when it comes to the extraction methods of starch and CNF used in F1, F2 and F3 [Table 3]. Although F1 exhibits exceptional mechanical properties to match HDPE's properties, its method could lead to pollution and environmental degradation. As a result, further research is needed to find alternative methods for the extraction methods included in this paper.

F1 is also the formula that overall mirrors the standards of conventional packaging. To incorporate this formula in large-scale manufacturing, although the sustainability standards would improve, its durability in stress conditions would require more testing. However, as an alternative material, it is feasible enough to be considered as benefitting the environment as not only is the material extracted from waste biomass that yields high mechanical properties, but it also degrades in the environment. This would allow for the starch-based, CNF-filled films to be used in packaging industries all over the world in the near future.

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