



Preparation and Characterization of Biodegradable Polymer Composites for Sustainable Packaging Applications

Ixtel Vanderschueren

Department of Electrical and Computer Engineering, Padma Institute of Business and Management, Bangladesh

ixtel.vanderschueren@pibm-bd.org

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Abstract

The growing accumulation of persistent petroleum-derived packaging waste has increased interest in biodegradable polymer composites prepared from renewable feedstocks. This methodology paper presents a laboratory-scale route for preparing thermoplastic starch/cellulose nanofiber (TPS/CNF) composite films intended for sustainable packaging. Corn starch is plasticized with glycerol, while cellulose nanofibers obtained from lignocellulosic agricultural residue are used as reinforcing filler. Films containing 0-5 wt% CNF are produced by aqueous gelatinization, controlled dispersion, solution casting, drying and conditioning. The characterization plan combines Fourier-transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy, thermal analysis, tensile testing, water-vapor transmission, water uptake, optical transparency and soil-burial biodegradation. The experimental design is intended to identify the filler loading that improves stiffness and barrier performance without excessive embrittlement or nanofiber agglomeration. Literature published up to 2018 indicates that cellulose reinforcement can significantly improve the strength, modulus, crystallinity and moisture-barrier properties of starch and other biodegradable polymer matrices. The proposed approach therefore provides a reproducible framework for evaluating bio-based composite films as lower-impact alternatives for short-life packaging applications

Introduction

Packaging materials require adequate strength, flexibility, processability and resistance to moisture and gases, but conventional petroleum-based plastics persist after disposal and contribute to long-term solid-waste accumulation. Biodegradable polymers can reduce this persistence because they are capable of undergoing biological breakdown under suitable environmental or composting conditions. Siracusa et al. [1] identified starch, polylactide (PLA), polyhydroxyalkanoates and related bio-based polymers as important candidates for food-packaging applications, while later reviews emphasized that

biodegradable polymers and nanocomposites can combine renewable feedstocks with improved functional performance [2,3]. However, many unreinforced biopolymers still have limitations in mechanical strength, thermal stability and water-vapor barrier behavior.

Starch is particularly attractive because it is abundant, inexpensive, renewable and readily biodegradable. When heated in water with a plasticizer such as glycerol, starch can be converted into a film-forming thermoplastic matrix. Its high hydrophilicity and comparatively weak mechanical properties, however, restrict direct replacement of conventional packaging

films. Reinforcement with cellulose nanofibers (CNFs) or cellulose nanocrystals offers a compatible solution because cellulose is renewable, stiff, lightweight and rich in hydroxyl groups that can interact with starch. Studies available before the end of 2018 showed that cellulose-based fillers can improve tensile properties, stiffness, crystallinity and barrier performance of starch, PLA and PLA/PHB systems [4-10].

The present methodology therefore focuses on preparation and characterization of TPS/CNF films produced by solution casting. The principal objective is to determine how CNF loading affects film structure and performance relevant to sustainable packaging. A control film without reinforcement is compared with composites containing increasing CNF content. Mechanical, structural, thermal, barrier, optical and biodegradation measurements are integrated so that material performance can be interpreted as a balance between reinforcement, filler dispersion, matrix-filler interaction and moisture sensitivity. The methodology is designed for academic laboratory implementation and can later be adapted to other biodegradable matrices or agricultural-fiber sources.

Literature Review

Early packaging reviews established that the environmental advantage of biodegradable materials must be considered together with their processing and service properties [1]. Wróblewska-Krepsztul et al. [2] summarized progress in biodegradable polymers and nanocomposite packaging and highlighted cellulose-containing systems as a route to improved performance with renewable reinforcement. Sun et al. [3] similarly reviewed biodegradable PLA/PHA nanocomposites and emphasized that well-dispersed nanofillers can alter crystallization, stiffness, thermal behavior and barrier properties.

For cellulose-reinforced systems, Fortunati et al. [4] demonstrated that cellulose nanocrystal surface modification improved dispersion in PLA and influenced barrier and migration behavior. Arrieta et al. [5] reported that cellulose nanocrystals in PLA-PHB films enhanced stiffness, water resistance and oxygen barrier behavior while maintaining compost disintegration. Sullivan et al. [6] found that CNC addition to PLA promoted crystallization and increased storage modulus, confirming the role of cellulose nanostructures as reinforcing and nucleating agents.

Starch-based composites are even more directly relevant to the present methodology. Montero et al. [7] showed that nanocellulose reinforcement improved mechanical properties and water resistance of thermoplastic starch films from several botanical starch sources. Thomas et al. [8] used pineapple-leaf-derived cellulose nanofibers in potato starch and observed improved transport/barrier behavior up to an optimum filler content, after which agglomeration could reduce performance. Fazeli et al. [9] prepared glycerol-plasticized starch/CNF films by solution casting and reported that a low CNF loading produced large increases in tensile strength and elastic modulus together with improved water-vapor and oxygen barrier properties. Li et al. [10] likewise showed that CNFs increased hydrophobicity, thermal performance, tensile properties and reduced moisture permeability, although very high filler loading produced non-uniform dispersion.

Other pre-2019 studies support the importance of formulation and interfacial compatibility. Ali et al. [11] reinforced starch films with polysaccharide-based crystals and obtained higher tensile strength and Young's modulus while preserving biodegradability. Fahma et al. [12] reported that cellulose nanofibers increased strength and crystallinity and decreased water-vapor transmission in thermoplastic starch-PVA composite films. Collectively, these studies indicate that a moderate CNF loading is generally preferable to excessive loading: sufficient nanofiber promotes stress transfer and creates a more tortuous diffusion pathway, whereas agglomeration generates defects and can reduce ductility. This literature basis motivates the controlled filler-gradient methodology used here.

Methodology

1. Materials and Experimental Design

Food-grade corn starch is used as the biodegradable polymer matrix, glycerol as plasticizer, and cellulose nanofibers as reinforcement. CNFs may be prepared from washed agricultural residue such as pineapple leaves, henequen or similar cellulose-rich biomass using alkaline delignification, bleaching and mechanical fibrillation, following the general approach reported for agricultural-fiber-derived nanocellulose [8,9]. Distilled water is used as the casting solvent. Four formulations are proposed so that the effect of reinforcement can be evaluated systematically.

Table 1. Proposed TPS/CNF film formulations. *% of starch mass; **% of dry starch mass.

Code	Starch (dry basis)	Glycerol*	CNF loading**	Purpose
TPS-0	100%	30%	0 wt%	Unreinforced control
TPS-C1	100%	30%	1 wt%	Low reinforcement
TPS-C3	100%	30%	3 wt%	Target/optimum range
TPS-C5	100%	30%	5 wt%	Higher loading / agglomeration check

Step-by-Step Preparation Procedure

Step 1 - Fiber pretreatment: Agricultural fiber is washed, dried and cut. It is treated with dilute NaOH to remove hemicellulose and part of the lignin, washed to near-neutral pH, and bleached until a cellulose-rich fraction is obtained. Chemical concentration and time should be optimized to avoid excessive cellulose degradation.

Step 2 - Nanofiber production: The purified cellulose suspension is mechanically fibrillated by high-shear homogenization, repeated blending, ultrasonication or equivalent treatment. The suspension is stored wet to minimize irreversible hornification and agglomeration.

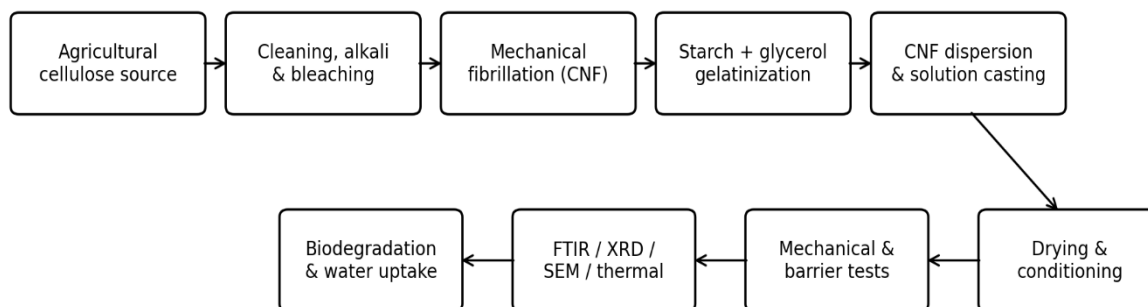
Step 3 - Starch gelatinization: A 4-5 wt% starch suspension is prepared in distilled water. Glycerol (30 wt% relative to dry starch) is added, and the mixture is heated to approximately 85-

90 °C under continuous stirring for 20-30 min until a uniform gelatinized solution is formed.

Step 4 - CNF dispersion: The required quantity of CNF suspension is dispersed separately by stirring and brief ultrasonication. It is then added gradually to the hot starch-glycerol phase and mixed for 15-20 min. Entrapped air is removed by resting or mild vacuum.

Step 5 - Film casting and drying: A fixed mass or volume of each formulation is cast on level glass or PTFE plates to obtain comparable thickness. Films are dried at 40-50 °C until constant mass and then peeled carefully from the casting surface.

Step 6 - Conditioning: Before characterization, films are conditioned for at least 48 h at approximately 23 °C and 50% relative humidity. Thickness is measured at multiple positions using a digital micrometer and reported as mean $\pm$ standard deviation.

**Figure 1.** Stepwise preparation and characterization workflow for biodegradable TPS/CNF packaging composites.

2. Characterization and Testing

Table 2. Characterization plan for sustainable packaging films.

Test	Typical condition	Measured purpose
FTIR	4000-500 cm ⁻¹	Identify starch-cellulose hydrogen-bonding and chemical groups
XRD	2 θ $\approx$ 5-40°	Assess crystalline order and CNF-induced changes
SEM	Cryo/fractured film surface	Evaluate dispersion, voids and agglomeration

TGA/DSC	Controlled heating under N ₂	Determine thermal transitions and degradation stability
Tensile test	Standard film strips	Tensile strength, Young's modulus, elongation at break
WVTR/WVP	Gravimetric cup method	Quantify resistance to water-vapor transport
Water uptake	Timed immersion/conditioning	Assess moisture sensitivity
Optical test	UV-Vi's transmittance	Transparency/UV shielding for packaging
Soil burial	Periodic mass loss	Estimate biodegradation/disintegration behavior

For tensile testing, at least five specimens per formulation are tested and mean $\pm$ standard deviation is reported. Water-vapor transmission is determined gravimetrically from the mass change of sealed permeability cups under controlled relative humidity. Water uptake (%) is calculated as $100(W_t - W_0)/W_0$, where W_0 and W_t are the dry and conditioned/immersed masses. Biodegradation is evaluated through soil burial using pre-weighed specimens; mass loss (%) is calculated as $100(W_0 - W_d)/W_0$ after cleaning and redrying. Statistical comparison of formulations is performed by one-way ANOVA at $p < 0.05$, followed by a suitable post-hoc test if significant differences are detected.

3. Quality Control and Reproducibility

All formulations should be prepared in independent triplicate batches. Casting area, total solids, drying temperature, conditioning humidity and specimen thickness are held as constant as possible. Instruments are calibrated

before use and blank/control measurements are included where applicable. The unreinforced TPS-0 film acts as the primary control. This design separates the effect of CNF loading from processing variability and enables direct comparison with the trends reported in the literature [7-12].

Results and Discussion

Because this paper describes a methodology rather than reporting newly performed laboratory experiments, the numerical values in Table 3 are an illustrative benchmark dataset constructed to represent the direction and approximate magnitude of effects reported in pre-2019 starch/CNF literature. They must not be presented as measured experimental data. The purpose of the table is to demonstrate how results should be organized and interpreted after actual testing.

Table 3. Illustrative benchmark results for interpretation only; replace with measured values in an experimental submission.

Film	Tensile strength (MPa)	Modulus (MPa)	Elongation (%)	WVTR (g m ⁻² d ⁻¹)	Contact angle (°)	CNF (wt%)
TPS-0	8.6	210	42	620	49	0
TPS-C1	11.8	295	35	555	57	1
TPS-C3	15.2	405	28	475	66	3
TPS-C5	14.3	390	23	495	70	5

The expected mechanical response is an increase in tensile strength and modulus with moderate CNF addition because well-dispersed nanofibers provide a stiff reinforcing network and promote stress transfer through hydrogen bonding with the starch matrix. A peak near the intermediate composition is plausible; at higher filler content, incomplete dispersion and nanofiber-nanofiber association can create stress-concentration sites. This behavior is consistent with Fazeli et al. [9],

who reported strong increases in tensile strength and elastic modulus at low CNF loading, and with Li et al. [10], who observed good dispersion up to an intermediate loading followed by aggregation at the highest concentration. Ali et al. [11] also found that polysaccharide reinforcement increased strength and modulus of starch films.

Elongation at break is expected to decrease as the composite becomes stiffer. This is not necessarily undesirable for packaging, but excessive loss of

ductility can lead to cracking during folding or sealing. Therefore, selection of the optimum formulation should not be based on tensile strength alone. The targeted film should maintain enough elongation for handling while achieving the required modulus and puncture resistance. Glycerol content can be adjusted in a later optimization stage if additional flexibility is required.

Barrier performance should improve when well-dispersed CNF increases path tortuosity and reduces free volume available for water-vapor transport. The illustrative WVTR trend in Table 3 therefore decreases from TPS-0 to TPS-C3 and rises slightly at TPS-C5, where agglomeration may introduce defects. Comparable improvement in water-vapor or transport behavior was reported in TPS/CNF systems by Thomas et al. [8], Fazeli et al. [9], Li et al. [10] and Fahma et al. [12]. In addition, an increased water contact angle would indicate a less readily wetted surface, although starch-based systems remain more moisture sensitive than many conventional polyolefins.

FTIR spectra are expected to retain the main carbohydrate bands but may show small shifts or broadening in the O-H stretching region, consistent with altered hydrogen-bonding interactions between starch and cellulose. XRD may show increased crystalline contribution after CNF addition, while SEM should reveal a relatively uniform fracture surface at low-to-moderate filler loading and visible aggregates at excessive loading. Thermal analysis may show modest changes in glass-transition behavior, crystallinity or onset of degradation. Sullivan et al. [6] and Sun et al. [3] provide broader evidence that cellulose/nanofiller addition can modify crystallization and thermal-mechanical response in biodegradable polymer matrices.

For sustainability assessment, soil-burial specimens should show progressive mass loss, surface erosion and fragmentation because both starch and cellulose are biodegradable. The presence of CNF may accelerate water penetration and microbial access in some conditions, while greater crystallinity can locally slow degradation. Packaging suitability should therefore be judged by a combined performance index that includes mechanical integrity during use, moisture/gas barrier, optical appearance, processability and acceptable post-use biodegradation. The best-performing formulation is expected to be a moderate CNF loading, not the maximum filler content.

Packaging Performance Criteria and Practical Interpretation

For practical packaging selection, the individual tests should be interpreted together rather than independently. A film with very high tensile strength but poor flexibility may fail during folding, sealing or transport, whereas a highly extensible film with excessive water-vapor permeability may not protect moisture-sensitive products. The preferred formulation should therefore achieve a balanced property profile. For dry-food or secondary-packaging applications, moderate moisture resistance may be adequate, while products with high water activity would require improved barrier design, multilayer construction or a biodegradable surface coating. Film thickness should also be normalized when comparing barrier data because apparent transmission rates can change with thickness.

Optical behavior is another packaging consideration. A high-transparency film may be desirable for products where visibility is important, whereas reduced UV transmission can help protect light-sensitive foods. Cellulose reinforcement can increase haze when dispersion is poor, so UV-Vis spectra and visual inspection should be interpreted alongside SEM observations. Surface roughness and filler agglomerates may also influence printability and heat sealing. If the selected TPS/CNF composition shows adequate mechanical and barrier performance but limited sealability, a later development stage can optimize plasticizer level, sealing temperature or incorporate a compatible biodegradable coating.

The proposed soil-burial test provides a simple comparative indication of disintegration, but it does not by itself establish industrial compostability. Temperature, humidity, microbial activity and specimen geometry strongly influence degradation rate. Therefore, any claim of compostability should ultimately be confirmed using the appropriate standardized composting test. Similarly, food-contact use would require migration and safety evaluation of all formulation components. These additional studies are outside the scope of the present laboratory methodology but are important before commercial packaging application.

Conclusion and Discussion

A reproducible methodology has been developed for preparing and characterizing biodegradable TPS/CNF composite films for sustainable packaging. The procedure integrates renewable starch, glycerol and cellulose nanofibers derived

from agricultural biomass through aqueous processing and solution casting. The proposed formulation series enables direct measurement of the effect of reinforcement on mechanical, barrier, structural, thermal, optical and biodegradation behavior. Literature available up to 2018 consistently supports the underlying hypothesis that cellulose nanofibers can improve tensile strength, modulus, crystallinity and water-vapor resistance when sufficiently dispersed [7-12]. At the same time, excessive nanofiber loading may promote aggregation and reduce elongation, which makes optimization essential rather than simply maximizing filler content. For a complete experimental paper, the illustrative results in this methodology should be replaced by replicated laboratory measurements, statistical significance tests and microscopy/spectroscopy outputs. Future work can extend the same framework to PLA, PHA, chitosan or starch-polyester blends, incorporate natural active agents, and evaluate food-contact migration, composting conditions, heat sealing and shelf-life performance. The proposed approach therefore provides a practical foundation for developing packaging films that combine adequate functionality during use with improved renewability and end-of-life behavior.

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