
Preliminary Results on Development of Sustainable and Biodegradable Food Packaging Films Using the Peels of Indigenous Banana Plant *Musa ingens* from Enga Province of Papua New Guinea

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Abstract: “Sustainability” and “Plastic/polymer pollution” are probably the two challenging phrases that the world is currently facing and the scientific investigations are focusing on. While mitigating plastic pollution has become a humongous task, sustainability has always come in handy with vastly spread biodiversity. Papua New Guinea (PNG) is one such blessed country with a hugely diversified bioresources. Food industry is one massive area that contributes to pollution, mostly through food packaging and single-use plastics. Of many known banana varieties in PNG, *Musa ingens* is special as it holds the record of being the world’s largest banana tree, and they are widely found at elevation limits between 1000-2100 m. Based on recent research published, this study was undertaken, however, with raw banana peels instead of ripened ones, to check if the fruit-ripening factor plays a significant role in preparing biofilms, and further if raw peels result in films with superior properties. The study indicated that raw peels were also able to produce biodegradable films. Preliminary results are encouraging and the films were able to be prepared, stretched as well as biodegraded within a definite timescale. The study indicated that some vital factors for putting these films into food packaging applications like water solubility (~21-47%, lower the better), moisture content (~16-35%, lower the better) and stretchability/elongation (~3-12%, higher the better) are still challenges to overcome. A discussion on subtle modifications done to the laboratory procedures and various results obtained will be presented. Further, a comparison of properties of films obtained from the present study with those of ripened peels reported in the literature will also be presented in the paper.

Keywords: Banana peels; *Musa ingens*; packaging films; biodegradable films; plastic pollution; sustainability.

1. INTRODUCTION

1.1 Plastics and Pollution

Plastics play an integrated role amongst the human community. Many different types of polymers are used in almost all areas of applications. They are widely being used due to their strength, manufacturability and workability, properties like unusual conductance, excellent biocompatibility, good insulation, to name a few. It is very unfortunate that most of the plastics and plastic additive chemicals do not effectively biodegrade, but rather decompose and transform, thus persisting in the environment and entering into atmosphere, hydrosphere, lithosphere and finally biosphere, where a high level of bioaccumulation and biomagnification result. In spite of the development of biodegradable and recyclable plastics, either that the littered plastics do not get biodegraded and decomposed within a short period of time, or do they reach proper recycling facilities to put them into reuse. Littered plastics, in the open environment, are constantly exposed to heat and light (sunlight), warmth (seawater and ground), microbes (land and water) and oxygen (atmosphere). Natural weathering of plastics results in embrittlement and breakdown to microplastics, and become very efficient in adsorbing metals and other persistent organic pollutants. Plastics degrade to micro- (<5 mm dia) and nano- (<1 µm dia) plastics and they get transported with the help of land and air and finally reach marine waterbodies wherein they stay for decades together.

1.2 Sustainable Solutions

Of all uses that plastics have been put into, packaging in food and beverage industries is one area where a wholesome of them are currently being used. Several sustainable solutions like use of lignocellulosic biomass are ventured into, but with a wide gap of essential functional properties like (i) resistance to degradation, moisture, puncture, temperature, gas and impact, (ii) impermeability to moisture, gas, oil and aromas, (iii) re-sealability and (iv) breathability. Vital characteristics like mechanical properties, shelf life, water vapour barrier and food decolouration and vitamin degradation are assessed using properties like (a) tensile strength, tear strength, elongation, (b) changes in moisture content and oxidative and microbial changes, (c) moisture loss and gain and (d) oxygen transmission rate and light transmission respectively. An estimate shows that 113 billion MT of carbon equivalent is generated in the earth by biomass, every year. Many cellulosic materials were used to prepare biopolymer films.

Papua New Guinea is a developing Pacific country belonging to Oceania continent. Based on the available statistics (10.73 million population, 65% falls between 15-64 years, ~45% of banana mass is peels, banana consumption rate in PNG 109 kg/capita/y, ~267 million kg of banana peels are produced annually in PNG which are littered and most of them undergo anaerobic degradation to produce methane gas, a strong greenhouse gas. By converting into biofilms, buildup of greenhouse gas is drastically reduced, simultaneously promoting sustainability and circular economy. In this short study, biofilms were prepared from unripe banana peels in order to evaluate their properties against those prepared from ripe bananas. Raw bananas from *Musa ingens* were used, which is the world's tallest banana tree, indigenous to PNG and grow in the highlands at an altitude of 1,000 to 2,100 m from the sea level. Table 1 shows the comparison of various parameters between ripe (RBP) and unripe banana peels (UBP). Peels from UBPs are found to be rich in cellulose, and hence chosen for the present study.

Table 1. Comparison of properties of RBP and UBPs.

#	Parameters	UBP (typical)	RBP (typical)
1	Moisture (%)	4.6 – 17.3	15.5 – 17.8
2	Protein (%)	1.94 – 2.41	2.18 – 2.73
3	Fat (%)	2.63 – 3.05	1.76 – 3.25
4	Ash (%)	11.9 – 13.5	11.3 – 14.7
5	Carbohydrates (%)	51.3 – 52.3	48.4 – 51.9
6	Energy (kJ/100g)	1015 - 1026	990 – 1012
7	Cellulose (%)	18 – 59	7 – 21
8	Hemicellulose (%)	17 – 20	6 – 20
9	Lignin (%)	7 - 16	6 – 12

2. EXPERIMENTAL

Banana peels were obtained from wild and unripe banana *Musa ingens* at lower Lai region of Wapenamanda district of Enga province, Papua New Guinea, with GPS coordinates of -5°40'0.012" N and 143°55'0.012" E. Laboratory grade reagents and deionized water were used throughout the study. Equipment like drying oven with natural convection (Binder, ED56 model), grinder (Brook Crompton, Series 2000 model), mechanical rotary shaker (Labec, TS-520D model), UV-visible spectrophotometer (Varian, Cary 50 Bio model) and centrifuging machine (Kokusen, H-103n model) were used. Subtle modifications to the reported procedure were followed for preparing biofilms (Fig.1, modifications shown in boxes above arrows). Formulae for calculating various parameters are shown in Equations (1)-(5), and the procedures followed were those reported in except for elongation test that was manually carried out.

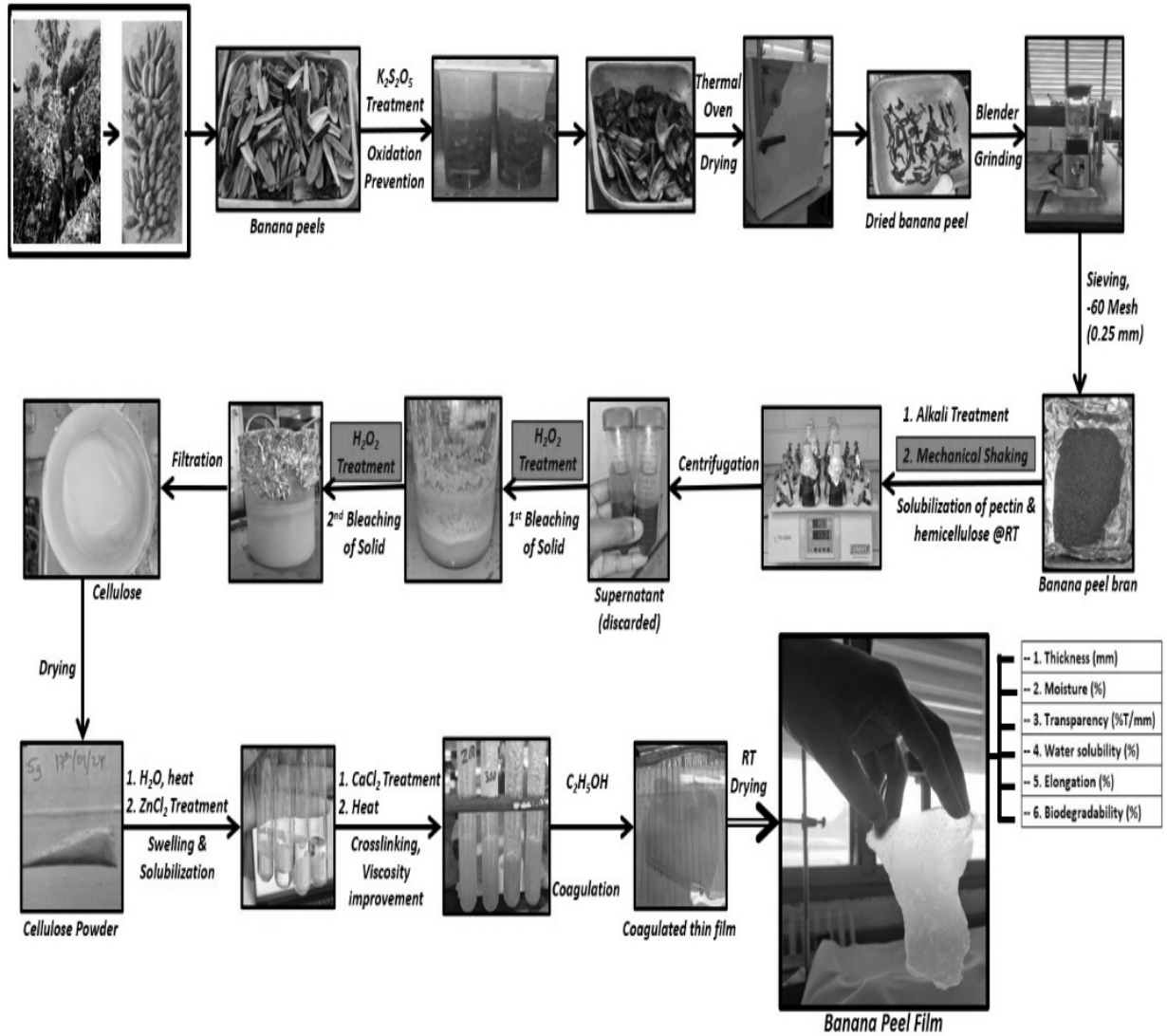


Fig. 1. Process flow chart for preparation of cellulose-based biofilms.

$$\text{Moisture content (\%)} = \left(\frac{\text{Initial Wt.} - \text{Final Wt.}}{\text{Initial Wt.}} \right) \times 100 \quad (1)$$

$$\text{Transparency (\%T/mm)} = \frac{\% \text{Transmittance @600 nm}}{\text{Film thickness (mm)}} \quad (2)$$

$$\text{Water solubility (\%)} = \left(\frac{\text{Initial Wt. of dry film} - \text{Wt. of insoluble portion after the test}}{\text{Initial Wt. of dry film}} \right) \times 100 \quad (3)$$

$$\text{Elongation (\%)} = \left(\frac{\text{Elongation at break (mm)} - \text{Initial length (mm)}}{\text{Elongation at break (mm)}} \right) \times 100 \quad (4)$$

$$\text{Biodegradability (\%area)} = \left(\frac{\text{Initial area of film (cm}^2\text{)} - \text{Area after 6 d of soil burrial (cm}^2\text{)}}{\text{Initial area of film (cm}^2\text{)}} \right) \times 100 \quad (5)$$

3. RESULTS

Measured properties of Raw/unripe banana peel films (UBPF) and comparison with reported values for those of RBP (RBPF) are shown in Table 2.

Table 2. Comparison of properties of UBPF, RBPF *V/s* polyethylene.

Description	[CaCl ₂], mM			
	200	300	400	500
<u>Film thickness (mm)</u>				
UBPF (this study)	0.03-0.08	0.04-0.05	0.03-0.04	##
RBPF	0.045-0.069	0.057-0.069	0.071-0.095	0.08-0.1
Polyethylene	----			
<u>Moisture content (%) ↓</u>				
UBPF (this study)	35.00	27.77	16.94 @	##
RBPF	22.30	19.57	18.12	13.73
Polyethylene	0.05-0.07			
<u>Transparency (%T/mm)</u>				
↑				
UBPF (this study)	26.23	34.30 \$	28.33 @	##
RBPF	45.10	38.35	34.82	31.97
Polyethylene	15-20			
<u>Water solubility (%) ↓</u>				
UBPF (this study)	46.66	37.00	21.00 @	##
RBPF	65.71	48.85	38.98	24.45
Polyethylene	0			
<u>Elongation (%) ↑</u>				
UBPF (this study)	12.0 @	8.2	3.8	##
RBPF	12.97	8.61	5.97	4.85
Polyethylene	69.33			
<u>Biodegradability (%) ↑</u>				
UBPF (this study), 6 days	100.00 @	84.12 \$	91.83	##
RBPF, 30 days	92.40	86.20	83.10	75.70
Polyethylene	0			

\$ - Probable error in measurements; @ - Preferable values for chosen parameters; ## - Not completed (study in progress); ↓↑ - Preferable trend for chosen parameters

Banana peel films prepared from varying Ca²⁺ concentrations (200, 300, 400 and 500 mM) are shown in Fig. 2.

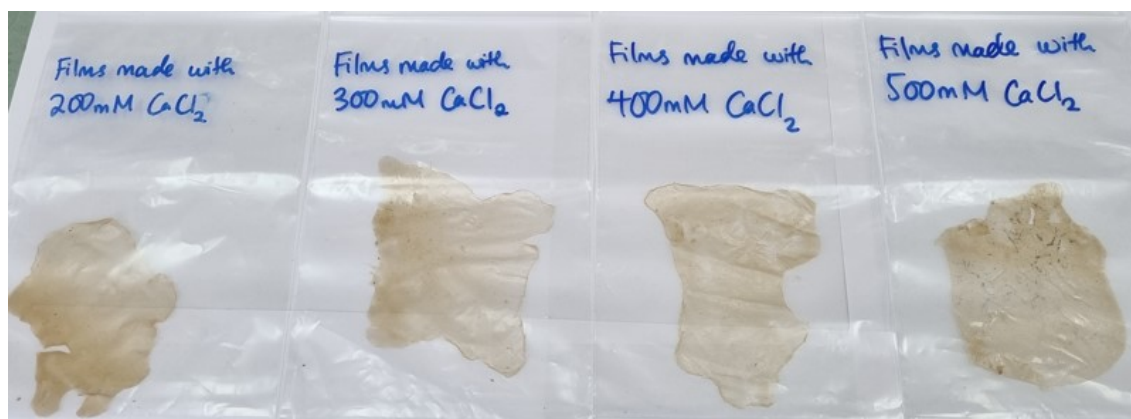


Fig. 2. BPF prepared using 200-500 mM CaCl₂ solutions.

4. DISCUSSION

4.1 Experimental Procedure

During the film preparation, alkali treatment was done in order to transform cellulose-I to its another polymorph (cellulose-II) which is proven to be thermodynamically more stable. Cellulose-II contains antiparallel chains of linked glucose units and intermolecular hydrogen bonds. A slight modification to the reported procedure was done by: (i) using relatively milder bleaching agent H_2O_2 to the insoluble residue instead of a stronger agent, $NaClO_2$, (ii) reducing the speed of mechanical rotary shaker from 350 rpm for 14 h to 200 rpm for 24 h, and (iii) reducing the speed of centrifuging machine from 10,000 rpm for 5 min. to 2,500 rpm for 20 min. Films were produced using differing $CaCl_2$ concentrations and 400 mM film was found to be clear and possessing easy film-forming tendency.

4.2 Properties of Banana Peel Films (BPF)

The measured thickness was found to have huge variations between various spots of the same film. This is due to the lack of thin layer chromatography applicator and hence the coagulated solid residue was manually spread on a flat surface. With higher concentrations of Ca^{2+} , properties like moisture content, water solubility, elongation% and biodegradability decrease. Polyethylene is a hydrophobic polymer with a very low moisture content compared to BPF prepared in the present study. This is due to the non-polar covalent bonds in polyethylene and higher degree of polar covalent bonds in the cellulosic structure. The lower transparency observed could be due to the use of milder bleaching agent compared to those published for RBPF. However, the trend with UBPF is found to be opposite when compared to RBPF, which warrants further investigation. Also, higher transparency observed in the present study compared to conventional polyethylene films is encouraging. Presence of other species due to ineffective bleaching has also probably influenced in reducing the water solubility. The films prepared in the present study are raw films with no addition of functional additives like plasticizers. Presence of higher $H_2O\%$ has significantly helped in imparting plasticizing effect, when plasticizers themselves are not present. This effect has necessarily helped the observed higher elongation. The decrease in biodegradability with increasing $[Ca^{2+}]$ could understandably be due to effective crosslinking of a number of Ca^{2+} ions between vertical chains providing certain matrix rigidity.

4.3 Mechanistic Aspects

The type of polymorph present in bio-resources is cellulose-1, that got converted to cellulose-II during the biofilm preparation. The original cellulose strands are hold together by weak van der Waals forces and H-bonds between hydroxyl groups (Fig. 3). When cellulose was treated with $ZnCl_2$, it solubilizes by breaking those weak bonds and coordinating with Zn^{2+} ions. The size of Zn^{2+} ions (88 pm) is insufficient to bridge two strands and hence single Zn^{2+} ion coordinates to a single strand. Later, when $CaCl_2$ was added, the relatively larger Ca^{2+} ions (114 pm) fill the gap and bridge two strands thus holding them together, allowing to improve the viscosity and enabling to coagulate from the solution. In order to enhance properties like elongation, flexibility, tear strength, etc. in bio-based films, a suitable MCl_x solution is required that improvises the current scenario with Ca^{2+} ions, which remains highly challenging. While strong covalent interactions persist in conventional polyethylene films (Fig. 4) that leads to properties like flexibility and stretchability, bio-based cellulosic films possess discrete ionic interactions between strands, that leads to limited flexibility.

Chemical guiding principles for further development process. (i) *Biocompatibility* – human bodies shall show no negative impact, for example, use of sodium salts might result in absorption of Na^+ ions from the film to the packed food, thus increasing human blood pressure levels; (ii) *nontoxic* – toxic heavy metal salts ($CdCl_2$, etc.) shall not be used as they are known to have biochemical toxic impacts after absorption; (iii) *ionic size* – shall best fit between strands such that a good stability is provided to impart strength, water vapour permeability, moisture absorption, etc.; (iv) *complex stability* – oxygen of -OH moieties interact with metal ions to form bonds; according to Pearson's hard soft acid base theory [18], $-O^-$ is a hard base and only suitable hard acid metal ions can interact

strongly to form stable interactions and hence stable films; and (v) *ionic charge* – plays a vital role in further stabilizing the coordination complex formed, in order to achieve desired properties.

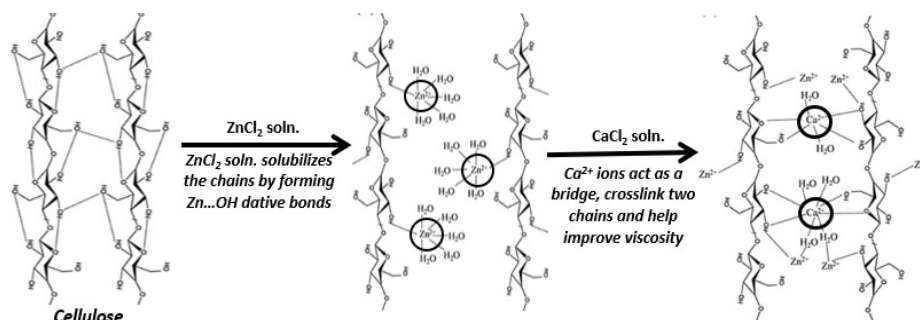


Fig. 3. Solubilizing and crosslinking in cellulose (strand structures adopted from [16]).

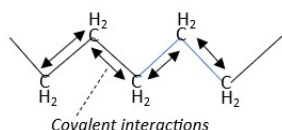


Fig. 4. Covalent interactions in conventional PE films.

Chemistry around calcium ions. The role of calcium ions in welan, an exopolysaccharide, was reported to bridge the two double helices through oxygens and water molecules, with metal's coordination number expanding to as high as 7-9 which are not so common, and further the $Ca^{2+} \dots O$ bond lengths varying from 2.61-3.42 Å. On a similar note, in this study, the role of calcium ions is also predicted to bridge cellulose strands which make the films strong enough, simultaneously imparting a limited flexibility. In the literature, positive charges of calcium ions were reported to be counterbalanced by two carboxylate units of the welan structure. However, in this study, since the previous experimental steps were not involved in alkali addition, the weakly acidic hydroxyl hydrogens (i.e., $org-OH \rightarrow org-O^- + H^+$) cannot be removed to counterbalance positive charges of Ca^{2+} ions.

4.4 Open Challenges

Several challenges persist in switching over from unsustainable to sustainable pathways. Some of them are: (a) meeting current demand *Vs* supply, (b) scaling up issues arise when switching over from a lab-scale preparation to massive industrial production, (c) structures of cellulose slightly vary from one to the other depending on origin (number of functional groups present, molecular weight, etc.), (d) matching of properties with current PE packaging films, and (e) many number of hydroxyl groups are present that affects properties like moisture content, water solubility, permeability to water vapour, etc., thus insisting on chemical modifications to cellulose structures by replacing hydrogens on -OH moieties with alkyl groups.

5. CONCLUSIONS AND FUTURE SCOPE

The study indicated that the properties of films derived from UBP match very well with RBP. However, extensive study on properties is required to proceed further. Biobased food packaging films are indeed a promising green alternative to polyethylene packaging films. Suitable physical, chemical or physicochemical modifications to the existing functionalities in banana peels need to be focused on to improve properties and hence their application potentials in industries, especially for food packaging. Some of the modifications like (i) surface coating to achieve suitable properties like water permeability, elongation and tensile strength, (ii) blending with suitable conventional plastics (in low percentages) or other bioplastic materials (like polylactic acid), or with other common plasticizers (like sorbitol, glycerol, etc.) can bring in essential characteristics like vapour-water barrier properties and stability, and (iii) chemical modifications on cellulosic part of banana peels can improve resistance to moisture uptake, tolerance to higher percentages of moisture, compatibility, etc. could perhaps maintain the freshness and tastes of

perishable foods (fruits, vegetables, seafoods, etc.). Thus, the reactivity / hydrophilicity / polarity of hydroxyl groups of cellulose can be modified using typical esterification and etherification reactions by converting hydroxyls to esters and alkoxy groups. While considering such modifications, it is worthy to focus on making the entire process “sustainable” and “renewable”. For example, property-enhancers like corn starch, carboxymethylcellulose, etc. could be derived from bio-sources which ultimately reduce waste and promote circular economy.

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