

Physico-Mechanical, Morphological and Thermal Analysis Of Bioplastic Films Fabricated from Sago Pith Waste Cellulose Fibres

Nurul Farhana Binti Zabidi

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Nurul Farhana Binti Zabidi

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DECLARATION

I declare that the work in this thesis was carried out in accordance with the regulations of Universiti Malaysia Sarawak. Except where due acknowledgements have been made, the work is that of the author alone. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Nurul Farhana Binti Zabidi

Signature	
Name:	Nurul Farhana Binti Zabidi
Matric No.:	19020125
Faculty of Engineering	
Universiti Malaysia Saraw	vak
Date:	5 th August 2024

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ABSTRACT

In light of the growing demand for sustainable alternatives to traditional plastics and the world's mounting environmental problems, this research explores the field of bioplastics with an emphasis on sago pith waste (SPW). The growing demand for biodegradable materials makes it essential to utilise resources such as SPW, which are plentiful in biomass, especially from the sago industry in Sarawak. The extraction of sago starch has led to significant environmental issues due to inadequate waste management practices. Therefore, while focusing on utilizing SPW, this research aims to fabricate a bioplastic film from SPW and investigate the optimal mass of cellulose fibres (CF) extracted from SPW solubilized in 20 mL of trifluoroacetic acid (TFA) to form a bioplastic film with good properties. First, to extract CF, the SPW underwent alkaline treatment and bleaching treatment. Then, CF was solubilized in TFA, with varying in mass from 0.10 g to 0.70 g. After the CF was fully dissolved, the process was then followed by solvent casting. The resulting bioplastic films produced were transparent with slight brown coloration. The SEM data revealed that bubble-shape protrusions on the bioplastic films appeared to shrink as the mass of CF decrease and a slight decrease in the number of visible pores were noted. Due to the improved morphological properties of the film, the tensile strength of BF-0.70 was enhanced by 90% as compared to BF-0.10. BF-0.70 also exhibited the best thermal stability as the main component of the bioplastic film started to degrade at 193°C and had the most amount of char residue. Out of all the other films, BF-0.70 also possessed the least water absorption ability, with a value of 86%, which was a good decrease from the 198% water absorption by BF-0.10. Thus, it was possible to determine the suitable CF mass. As evidenced by the features of BF-0.70, the results had proven that 0.70 g is the

optimal mass of CF that can be solubilised in TFA to form a bioplastic film with good properties.

Keywords: Sago pith waste, TFA, alkaline treatment, solvent casting, FTIR, water absorption

Analisis Fiziko-Mekanikal, Morfologi dan Termal Filem Bioplastik yang Dihasilkan daripada Serat Selulosa Hampas Sagu

ABSTRAK

Dalam usaha untuk memenuhi permintaan yang semakin meningkat terhadap alternatif yang lestari kepada plastik tradisional dan masalah alam sekitar yang semakin serius, kajian ini meneroka bidang bioplastik dengan penekanan kepada hampas sagu. Permintaan yang meningkat terhadap bahan biodegradasi menjadikan penggunaan sumber seperti hampas sagu, yang banyak terdapat dalam biojisim, amat penting terutamanya dari industri sagu di Sarawak. Pengekstrakan kanji sagu telah menyebabkan isu alam sekitar yang ketara akibat daripada amalan pengurusan sisa yang tidak lengkap. Oleh itu, dengan memberi tumpuan kepada penggunaan hampas sagu, kajian ini bertujuan untuk menghasilkan filem bioplastik daripada hampas sagu dan menyiasat jisim serat selulosa yang optimum diekstrak daripada hampas sagu yang dilarutkan dalam 20 mL asid trifluoroasetik (TFA) untuk membentuk filem bioplastik dengan sifat yang cemerlang. Pertama sekali, untuk mengekstrak serat selulosa, SPW telah melalui rawatan alkali dan rawatan pelunturan. Kemudian, serat selulosa dilarutkan dalam TFA, dengan variasi jisim dari 0.10 g hingga 0.70 g. Setelah serat selulosa larut sepenuhnya, proses diteruskan dengan pengacuan pelarut. Filem bioplastik yang terhasil adalah lutsinar dengan sedikit pewarnaan coklat. Data SEM mendedahkan bahawa bonjolan berbentuk gelembung pada filem bioplastik kelihatan mengecut apabila jisim serat selulosa berkurangan dan sedikit pengurangan dalam bilangan liang yang kelihatan telah diperhatikan. Disebabkan oleh penambahbaikan sifat morfologi filem tersebut, kekuatan tegangan BF-0.70 meningkat sebanyak 90% berbanding dengan BF-0.10. BF-0.70 juga mempamerkan kestabilan terma terbaik kerana komponen utama filem bioplastik mula terurai pada 193°C dan mempunyai

sisa arang paling banyak. Daripada semua filem lain, BF-0.70 juga mempunyai kebolehan penyerapan air yang paling rendah, dengan nilai 86%, yang merupakan penurunan yang baik daripada penyerapan air sebanyak 198% oleh BF-0.10. Oleh itu, jisim serat selulosa yang terbaik telah dijumpai. Seperti yang dibuktikan oleh ciri-ciri BF-0.70, keputusan telah membuktikan bahawa 0.70 g adalah jisim serat selulosa yang boleh dilarutkan dalam TFA untuk membentuk filem bioplastik dengan sifat yang baik.

Kata kunci: Hampas sagu, TFA, rawatan alkali, FTIR, pencelupan pelarut, keupayaan penyerapan air

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LIST OF ABBREVIATIONS

BF	Bioplastic film	
BPA	Bisphenol A	
CF	Cellulose fibres	
CGS	Centre for Graduate Studies	
CRAUN	Crop Research and Application Unit	
DoA	Department of Agriculture	
EDX	Energy dispersive X-ray	
EMBL-EBI	European Molecular Biology Laboratory–European	
	Bioinformatics Institute	
FTIR	Fourier transform infrared	
FTIR-ATR	Fourier transform infrared attenuated total reflectance	
GDP	Gross domestic product	
HDPE	Polyethylene high density	
LDPE	Polyethylene low density	
MCC	Microcrystalline cellulose	
NOAA	National Oceanic and Atmospheric Administration	
OECD	Organisation for Economic Co-operation and Development	
OSL	Ocean surface layer	
PHA	Polyhydroxyalkanoates	
PLA	Polylactic acid	
PP	Polypropylene	
PS	Polystyrene	
PVC	Poly(vinyl) chloride	

SEM	Scanning electron microscope
SPW	Sago pith waste
SSCMF	Sago starch filled with cellulose microfibres
TFA	Trifluoroacetic acid
TFAA	Trifluoroacetic anhydride
TGA	Thermogravimetric analysis
TS	Tensile strength
UN	United Nations
UNIMAS	Universiti Malaysia Sarawak

CHAPTER 1

INTRODUCTION

1.1 Study Background

The consumption of fossil fuels for energy and manufacturing processes has led to the rapid depletion of these non-renewable resources, significantly impacting the environment. Today, petroleum is the main source of plastic, with approximately onefourth of each barrel of oil used to produce plastics and other goods (Marrone, 2022). In 2022, over 390 million tonnes of fossil fuel-based plastics were manufactured, contributing to a severe global plastic pollution problem (Ali et al., 2023). This pollution poses serious threats to both human and ecological health, necessitating the urgent need for sustainable alternatives.

In response to these challenges, the development of sustainable materials such as bioplastics has gained considerable attention. Bioplastics, derived from renewable biomass sources, offer a promising alternative to conventional plastics made from fossil fuels (Filho et al., 2022). They not only reduce the dependency on finite fossil resources but also provide potential solutions for waste management issues. Some bioplastics are highly biodegradable, contributing to more environmentally sustainable practices and reducing the environmental footprint associated with plastic waste (Lamberti et al., 2020). Implementing bioplastics could significantly mitigate soil, water, and air pollution, aligning with global efforts to adopt more sustainable and eco-friendly practices.

Despite their potential, the production of bioplastics faces several obstacles, primarily due to higher production costs, which are two to three times more expensive than traditional plastics and has lower production yields. Additionally, the manufacturing process encounters various difficulties. For example, some bioplastics possess inferior mechanical properties, making them more brittle and prone to tearing compared to oil-based plastics. Furthermore, bioplastics often have weaker barrier properties than conventional plastics, resulting in a shorter lifespan. Despite these challenges, ongoing efforts aim to enhance the production processes and mechanical properties of bioplastics, making them a more feasible alternative to conventional plastics (Ibrahim et al., 2021).

One such effort involves creating bioplastics from agricultural waste such as sago pith waste (SPW). The utilization of SPW as a raw material for bioplastic production holds great promise in addressing both waste management and sustainable resource utilization. Nevertheless, raw SPW cannot be used to produce higher value-added materials because of the composition of the waste. Previous research demonstrated that starch had been extracted from SPW with the assistance of ultrasound to form bioplastic (Tan et al., 2021) and CF can be extracted from treating SPW through alkaline treatment and bleaching treatment. Yacob and his team treated SPW by using alkaline and bleaching treatment to extract CF to act as a reinforcing agent, specifically to improve the properties of starch films produced in the study (Yacob et al., 2018). Meanwhile, Jampi and her team also used similar techniques to extract CF from SPW, before preparing it into hydrogel (Jampi et al., 2021).

Building on these findings, treatments to extract CF were applied in this study, as past research has used cellulose-based raw materials to form bioplastic films by solubilizing them in trifluoroacetic acid (TFA) and solvent casting (Bilo et al., 2018). A method discovered by Bayer and his team uses trifluoroacetic acid (TFA) as a solvent to solubilize cellulose and similar compounds as well as several agricultural plant wastes to form bioplastic films (Tedeschi et al., 2020). TFA aids in the formation of a homogenous solution that can be cast into a bioplastic film. Due to its ability to swell cellulose effectively and its ability to be recycled because of its low boiling point, trifluoroacetic acid (TFA) is of interest since it offers an effective method to reduce cellulosic crystallinity and break up hydrogen bonds within the crystalline region (Liu et al., 2021)

While TFA has shown promise in creating bioplastics from various sources, such as rice hulls, green herbs, cocoa pod husks, and seaweed (Farid et al., 2021), a substantial research gap persists. Currently, there is no research that uses CF extracted from SPW to produce bioplastic films by using TFA. Furthermore, there is a lack of investigation into identifying the mass of cellulose fibres (CF) needed from SPW in a fixed volume of TFA to form a bioplastic film. Consequently, the influence of CF mass on critical properties of resulting bioplastic films including morphology, mechanical strength, thermal characteristics, and water absorption remains unexplored. Addressing this gap is crucial for optimizing the production process and enhancing the overall performance of bioplastic films.

Therefore, the research focuses on addressing this research gap by investigating the effects of the different masses of CF in a fixed volume of TFA on the properties of the resulting bioplastic films. In addition, the FTIR, morphological, mechanical, and thermal properties as well as water absorption of bioplastic films produced were also analysed.

1.2 Problem Statement

The main problem of the research is the significant environmental challenges posed by the improper waste management of SPW generated from sago starch extraction in Sarawak, where 90% of the world's sago starch is produced. Despite the abundance of SPW, its inefficient handling leads to severe water and air pollution. This research addresses the need for sustainable solutions by focusing on the extraction and utilization of CF from SPW to produce bioplastic films. The critical research gap lies in investigating the mass of CF that can be solubilized in a fixed volume of TFA to enhance the morphological, mechanical, and thermal properties, as well as water absorption capabilities, of the resulting bioplastic films.

1.3 Hypothesis

The combination of alkali and bleaching treatments of SPW will effectively extract CF suitable for bioplastic film formation. It is hypothesized that the mass of CF solubilized in 20 mL of TFA will significantly influence the properties of the resulting bioplastic films. Among the varying masses of CF, 0.70 g of CF is expected to yield a bioplastic film with good properties, including enhanced tensile strength, improved thermal stability, and reduced water absorption.

1.4 **Objectives**

The research's objectives include:

i. To extract cellulose fibres from SPW for bioplastic production.

ii. To investigate the mass of CF used in forming bioplastic films per 20 mL of TFA.

1.5 Scope of Study

This study focuses on the sustainable utilization of SPW from the sago industry in Sarawak to address significant environmental challenges caused by improper waste management. The scope of the research encompasses the following key areas: i. Extraction of Cellulose Fibres (CF):

Implementing alkali and bleaching treatments on SPW to extract CF, transforming waste into a valuable raw material suitable for bioplastic film production.

ii. Solubilization and Bioplastic Film Formation:

Investigating the solubilization of CF in TFA at varying masses (0.10 g to 0.70 g) to form bioplastic films as well as utilizing solvent casting to produce bioplastic films and examining their physical appearance.

iii. Characterization of Bioplastic Films:

a. Performing Fourier Transform Infrared Spectroscopy (FTIR) analysis to study chemical compositions and functional groups present in the bioplastic films.

 b. Conducting morphological analysis using Scanning Electron Microscopy (SEM) to observe structural properties and pore distribution.

c. Evaluating mechanical properties such as tensile strength to determine the film's durability and robustness.

d. Assessing thermal stability through thermal degradation tests to determine the film's resistance to heat.

e. Measuring water absorption capacity to understand the film's potential for applications requiring moisture resistance.

This research hopes to contribute to sustainable waste management practices by converting SPW into high-value bioplastic films, thus mitigating environmental pollution and promoting the development of biodegradable materials.

CHAPTER 2

LITERATURE REVIEW

2.1 Polymers

A polymer is a material, either natural or synthetic, made up of extremely large molecules called macromolecules. These macromolecules are formed by linking together many smaller units called monomers. Polymers are abundant in biological organisms and include substances such as proteins, cellulose, and nucleic acids (Rial, 2022).

2.1.1 Types of Polymers

Based on their synthesis, polymers can be classified into two types: synthetic and natural as discussed in Chapter 2.2 and 2.3.

2.2 Synthetic Polymers

Synthetic polymers are built with long chains of atoms and put together in repeating units, usually way longer than the polymers that occur in nature. Therefore, synthetic polymer is sturdy, lightweight, and elastic due to the length of the chains and their arrayed patterns. Synthetic polymers are very useful due to their properties, causing excellent growth of in the production of plastic, beating many other man-made materials (Davoodi et al., 2024).

The word plastic originally meant "pliable and easily shaped". Humans have learned to manufacture plastic over 150 years ago, regularly using the plentiful carbon atoms obtained from petroleum and fossil fuel (Davoodi et al., 2024). Plastic is an amazing material that stimulates economic growth and boosts modern advancements through its synthetic properties. They are a valuable resource that offer numerous advantages to civilisation, including improving their well-being, cleanliness, and safety. Moreover, plastic materials have played a vital role in multiple industries, such as food packaging, drug delivery, fuel production, disease prevention, and road construction. In 2019, the packaging sector had dominated the plastic market, with building and construction, textiles, automotive and transportation, infrastructure and construction, and consumer goods tailing closely behind (Kumar et al., 2021).

2.2.1 Addition Polymers

Plastics are normally made from addition polymers, as it results in strong and durable materials that can be moulded into a wide range of shapes and sizes. Addition polymerization, which is also known as chain reaction polymerization, necessitate that the monomers possess no less than one double bond. Both molecules and by-products are neither removed nor created in this type of polymerization (Youssef, 2022).

The monomers had been joined through addition reaction and no atoms have been lost (Youssef, 2022). Additionally, most polymers are composed of many different types of molecules such as thousands of monomer units that can be found along HDPE molecules. The common addition polymers can be observed in Table 2.1.