



UNIVERSITI PUTRA MALAYSIA

DEVELOPMENT AND CHARACTERIZATION OF ARROWROOT (*Maranta arundinacea* L.) FIBRE-REINFORCED THERMOPLASTIC STARCH BIOCOMPOSITES

TARIQUE JAMAL

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By

TARIQUE JAMAL

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Doctor of Philosophy

July 2022

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DEDICATION

To Al-Quran, the greatest source of knowledge

“Such as remember Allah, standing, sitting, and reclining, and consider the creation of the heavens and the earth, (and say): Our Lord! You created not this in vain. Glory be to You! Preserve us from the doom of Fire” (Surah Al-Imran 3: 191)

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To my beloved father and mother

&

To my beloved grandfather and grandmother

&

To my awesome siblings

&

To my beloved wife

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

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July 2022

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Faculty : Engineering**

Petroleum-based plastics are one of the most commonly used materials in the packaging industry, which has been a source of concern for the global environment. The increased production of harmful environmental plastic waste has fueled the development of natural-based, renewable, and biodegradable materials. Therefore, environmental sustainability and the absence of harmful carbon emissions during and after processing are desirable characteristics of potential materials. Furthermore, the widespread acceptance of natural fibres and biopolymers as green materials is being driven by the rapid depletion of petroleum resources, as well as a growing awareness of global environmental issues associated with the use of conventional plastics. Also, rising petrochemical prices and environmental concerns are driving the development of natural polymeric materials for a wide range of applications in food-packaging materials that are more consumer-friendly. Among these materials, arrowroot (*Maranta arundinacea* L.) has emerged as a vital and effective source of starch and fibres. Arrowroot belongs to the Marantaceae family, which is typically found in tropical forests. When compared to other agro-based products, arrowroot starch and fibres has some distinct advantages, including lower cost than other natural sources and greater accessibility. Therefore, several laboratory experiments were conducted to produce and characterize arrowroot fibres, biopolymers, and biocomposite films. The samples were developed using a solution casting method. Initially, arrowroot bagasse (ABF) and husk fibres (AHF) were extracted and the physical, chemical, thermal, morphological properties, as well as crystallinity, were characterized. The chemical composition analysis revealed that ABF has higher cellulose (45.97 %) than AHF (37.35 %), cassava bagasse (10.04 %), and corn hull (15.30 %). In addition, ABF is significantly low in lignin (2.78 %) and density (1.11 g/cm³) than AHF, corn hull, and cassava. Concerning the above characterization of fibres, it was found that the lignocellulosic biomasses from arrowroot are alternative promising sustainable material, which can be used in food packaging as a renewable filler.

The second stage was designed to investigate the development of arrowroot starch (AS) films using glycerol (G) as a plasticizer at the ratio of 15, 30, and 45% (w/w, starch basis) to achieve a new biopolymer for the application of environmentally friendly materials. The developed films were analysed in terms of physical, structural, mechanical, thermal, environmental, and barrier properties. The incorporation of glycerol into AS film-making solution reduced the brittleness and fragility of films. An increment in glycerol concentration caused an increment in film thickness, moisture content, and solubility in water, whereas density and water absorption were reduced. The tensile strength and modulus of G-plasticized AS films were reduced significantly from 9.34 to 1.95 MPa and 620.79 to 36.08 MPa, respectively, while elongation at break was enhanced from 2.41 to 57.33 %. FTIR analysis revealed that intermolecular hydrogen bonding occurred between glycerol and AS in plasticized films compared to control films. The G-plasticized films showed higher thermal stability than control films. Water vapour permeability (WVP) of plasticized films increased by an increase in glycerol concentrations. Furthermore, a novel biodegradable thermoplastic arrowroot starch (TPAS) film containing arrowroot fibre (AF) at different concentrations (0, 2, 4, 6, 8, and 10 wt.%) was developed and characterized in terms of thermal, antibacterial activity, water vapor permeability (WVP), biodegradability, physical, morphological (FESEM), tensile and tear strength, and light transmittance properties. The TPAS/AF biocomposite film revealed a higher degradation temperature (313.02 °C) than other biocomposite films, indicating better thermal stability. Furthermore, increasing AF concentration led to a significant ($p < 0.05$) reduction in the linear burning rate and WVP of the biocomposite films from 248.9 to 115.2 mm/min and $8.18 \times 10^{-10} \times \text{g. s}^{-1} \cdot \text{m}^{-1} \cdot \text{Pa}^{-1}$ to $5.20 \times 10^{-10} \times \text{g. s}^{-1} \cdot \text{m}^{-1} \cdot \text{Pa}^{-1}$, respectively. The tensile and tear strengths of TPAS/AF composites were increased significantly from 2.42 to 15.22 MPa and 0.83 to 1.28 MPa, respectively, and the elongation was decreased from 46.62 to 6.21%. The findings revealed that after being reinforced with fibres, the mechanical properties enhanced, and the optimum filler content was 10%. Regardless of fibre loadings, the results of water absorption testing revealed that the composite films immersed in seawater and rainwater absorbed more water than distilled water. In addition, the incorporation of AF and control film showed an insignificant effect against three pathogenic bacteria including *Staphylococcus aureus* (ATCC 43300), *Escherichia coli* (ATCC 25922), and *Bacillus subtilis* (B29). The soil burial findings demonstrated that the weight loss of TPAS/AF biocomposite films was significantly higher than TPAS film. Overall, the reinforcement of arrowroot fibre with TPAS film improves the properties of biocomposites for environmentally friendly food packaging applications. The development of fully biodegradable packaging films is essential in the continuous effort to address current environmental issues and gradually replace commonly used conventional packaging materials.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

**PEMBANGUNAN DAN PENCIRIAN BIOKOMPOSIT KANJI
TERMOPLASTIK DIPERKUAT GENTIAN UBI BEMBAN (*Maranta
arundinacea* L.)**

Oleh

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Plastik berasaskan petroleum adalah salah satu bahan yang digunakan dalam industri pembungkusan, dan ia telah menjadi kebimbangan terhadap persekitaran global. Peningkatan pengeluaran sisa plastik yang amat berbahaya kepada alam sekitar telah memacu pembangunan bahan berasaskan semula jadi, yang boleh diperbaharui dan terbiodegradasi. Oleh itu, kemampanan alam sekitar dan ketiadaan pelepasan karbon berbahaya semasa dan selepas pemrosesan adalah ciri-ciri yang diinginkan bagi bahan berpotensi. Tambahan pula, penerimaan meluas terhadap gentian semula jadi dan biopolimer sebagai bahan hijau didorong oleh penyusutan pesat sumber petroleum, serta kesedaran yang semakin meningkat tentang isu alam sekitar global yang berkaitan dengan penggunaan plastik konvensional. Di antara bahan-bahan ini, ubi bemban atau lebih dikenali sebagai ubi bemban (*Maranta arundinacea* L.) telah muncul sebagai sumber kanji dan serat yang penting dan berkesan. Ubi bemban tergolong dalam keluarga *Marantaceae*, yang biasanya ditemui di hutan tropika. Sampel telah dibangunkan menggunakan kaedah tuangan larutan. Pada mulanya, gentian ampas ubi bemban (ABF) dan gentian sekam ubi bemban (AHF) diekstrak, seterusnya dicirikan dari segi sifat fizikal, kimia, haba, morfologi, serta kehabluran ABF dan AHF. Analisis komposisi kimia menunjukkan bahawa ABF mempunyai selulosa yang lebih tinggi (45.97%) daripada AHF (37.35%). Tambahan pula, kandungan lignin (2.78%) dan ketumpatan (1.11g/cm^3) ABF adalah ketara rendah berbanding dengan AHF, kulit jagung dan ubi kayu. Mengenai pencirian gentian di atas, didapati bahawa biojisim lignoselulosa daripada akar ubi bemban merupakan bahan mampan alternatif yang menjanjikan, yang boleh digunakan dalam pembungkusan makanan sebagai pengisi boleh diperbaharui.

Pada peringkat kedua pencirian filem kanji ubi bemban (AS) dijalankan menggunakan gliserol (G) sebagai pemplastik pada nisbah 15, 30, dan 45% (b/b, asas kanji). Filem yang dibangunkan telah dianalisis dari segi sifat fizikal, struktur, mekanikal, dan haba.. Penggabungan gliserol ke dalam pembuatan tuangan larutan filem AS mengurangkan kerapuhan, ketumpatan dan penyerapan air filem manakala ketebalan, kandungan

lembapan dan keterlarutan dalam air meningkat. Kekuatan tegangan dan modulus filem AS yang diplastik-G telah berkurangan dengan ketara daripada 9.34 kepada 1.95 MPa dan 620.79 kepada 36.08 MPa, masing-masing, manakala pemanjangan semasa putus meningkat daripada 2.41 kepada 57.33%. Analisis FTIR mendedahkan bahawa ikatan hidrogen antara molekul berlaku antara gliserol dan AS dalam filem plastik berbanding filem kawalan. Tambahan pula, filem kanji ubi bemban (TPAS) termoplastik terbiodegradasi novel yang mengandungi serat ubi bemban (AF) pada kepekatan berbeza (0, 2, 4, 6, 8, dan 10 wt.%) telah dibangunkan dan dicirikan dari segi haba, aktiviti antibakteria, kebolehtelapan wap air (WVP), kebolehibiodegradan, fizikal, morfologi (FESEM), kekuatan tegangan dan koyak, dan sifat penghantaran cahaya. Filem biokomposit TPAS/AF mendedahkan suhu degradasi yang lebih tinggi (313.02 °C) berbanding filem biokomposit yang lain, menunjukkan kestabilan terma yang lebih baik. Tambahan pula, peningkatan kepekatan AF membawa kepada pengurangan yang ketara ($p < 0.05$) dalam kadar pembakaran linear dan WVP bagi filem biokomposit daripada 248.9 kepada 115.2 mm/min dan $8.18 \times 10^{-10} \times \text{g} \cdot \text{s}^{-1} \cdot \text{m}^{-1} \cdot \text{Pa}^{-1}$ hingga $5.20 \times 10^{-10} \times \text{g} \cdot \text{s}^{-1} \cdot \text{m}^{-1} \cdot \text{Pa}^{-1}$, masing-masing. Kekuatan tegangan dan koyak komposit TPAS/AF telah meningkat dengan ketara daripada 2.42 kepada 15.22 MPa dan 0.83 kepada 1.28 MPa, dan pemanjangan telah menurun daripada 46.62 kepada 6.21%. Penemuan mendedahkan bahawa selepas diperkukuh dengan gentian, sifat mekanikal dipertingkatkan, dan kandungan pengisi optimum ialah 10%. Tanpa mengira beban gentian, keputusan ujian penyerapan air mendedahkan bahawa filem komposit yang direndam dalam air laut dan air hujan menyerap lebih banyak air daripada air suling. Di samping itu, penggabungan AF dan filem kawalan menunjukkan kesan yang tidak ketara terhadap tiga bakteria patogen termasuk *Staphylococcus aureus* (ATCC 43300), *Escherichia coli* (ATCC 25922), dan *Bacillus subtilis* (B29). Penemuan daripada ujian biodegradabiliti menunjukkan bahawa kehilangan berat filem biokomposit TPAS/AF adalah lebih tinggi daripada filem TPAS. Secara keseluruhannya, pengukuhan gentian ubi bemban dengan filem TPAS meningkatkan sifat biokomposit untuk aplikasi pembungkusan makanan mesra alam.

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LIST OF SYMBOLS

$\%$	Percentage
μm	Micrometer
g	Gram
h	Hour
M_f	Weight final
M_i	Weight initial
mm	Milimeter
MPa	Mega pascal
$^\circ$	Degree
$^\circ C$	Degree celcius
T_g	Glass transition
T_{Onset}	Onset temperature
T_p	Peak temperature
T_r	Light transmittance
V	Volume
W_L	Weight loss
$wt.\%$	Weight percentage
Δm	Weight difference
ρ	Density

LIST OF ABBREVIATIONS

ABF	Arrowroot bagasse fibre
ADF	Acid detergent fibre
AF	Arrowroot fibre
AHF	Arrowroot husk fibre
ANOVA	Analysis Of Variance
AS	Arrowroot starch
ASTM	American Society for Testing and Materials
DSC	Differential Scanning Calorimetry
DTG	Derivative thermogravimetry
FESEM	Field emission scanning electron microscopy
FTIR	Fourier Transform Infrared
G	Glycerol
NDF	Neutral detergent fibre
SEM	Scanning Electron Microscopy
TGA	Thermal-gravimetric analysis
TPAS	Thermoplastic arrowroot starch
TS	Tensile strength
UTM	Universal testing machine
WVP	Water vapor permeability
XRD	X-ray diffraction

CHAPTER 1

INTRODUCTION

1.1 Background

The abundance of petroleum-based waste disposal has significantly contributed to rising environmental pollution, causing problems for both natural life as well as human health. Plastic's invention escorted in a revolution in material production in a variety of industries, including medicine, automobiles, electronics, and packaging (Sharuddin et al., 2016). It is distinguished by its reliability, heat resistance, and appropriateness for large scale production. The global production of synthetic plastics reached 140 million tons annually, an increase of 2% per year (Siracusa, 2019). Between 1950 and 2017, an estimated 9,200 million metric tons of synthetic plastics were produced, with 5,300 metric tons entering landfills, dumps, or the environment (Gavigan et al., 2020). This demonstrates that most of the plastic rubbish that landed up in landfills in extremely high and takes up a significant amount of space. Because plastics are certainly valuable and useful in our daily lives, several material engineers and researchers are attempting to design plastic materials that are both safer and much better for the environment. Bioplastics, which are derived from plant crops instead of fossil fuels, are being developed by manufactures as a more environmentally friendly substitute for petroleum-based plastics. Some scientists are trying to find a way to make recycling more efficient, with the goal of optimizing the process of transforming plastics back into the fossil fuels that they came from. All these researchers understand that plastics are not ideal, but an essential and vital component of our present and future applications (Rhim et al., 2013). These issues have motivated researchers and scientists to design and develop environmentally friendly advanced materials produced from renewable resources to substitute non-biodegradable materials in a variety of applications, protecting the green environmental. Among these source materials, arrowroot biopolymers and biocomposites offer numerous benefits, including outstanding reliability, low cost, availability, biodegradability, as well as high content of starch.

Arrowroot (*Maranta arundinacea* L.) is a large perennial herb native to the tropical forest that belongs to the *Marantaceae* family. The arrowroot plant is primarily found in the West Indies (Jamaica), Brazil, Indonesia, Malaysia, Philippines, India, and Sri Lanka (Nogueira et al., 2018). Arrowroot starch has excellent properties such as digestibility and gelling ability, and the highest amylose content (40.86%) (Gordillo et al., 2014), competing with corn starch (28-33%), wheat starch (30-32%), potato (18-20%), and cassava starch (16-19%), all of which are required for film production. Previous research has shown that the amylose content of starch influences its film-forming properties; hydrogen bonding linear chains together forms strong and stiff films. As a result, the high amylose content of arrowroot starch produces stronger films than other starch sources. A significant amount of waste arrowroot fibres is obtained during the arrowroot starch extraction process. According to a literature, arrowroot rhizomes comprise 38.1% of bagasse fibre (Branco et al., 2019). Branco et al. (2019) found that the arrowroot bagasse fibres are coarser as well as longer in comparison to cassava bagasse fibres. Biopolymer films were made from arrowroot starch employing a solution casting process

with 15 to 45% (by weight) glycerol as a plasticizer. The results revealed that incorporating 30 % glycerol into the biopolymer was the most effective glycerol concentration, resulted in outstanding thermal and physical properties. Despite having adequate characteristics, the developed biopolymer has certain drawbacks, significantly such as poor tensile and water resistance. Hence, to overwhelm such drawbacks, arrowroot starch reinforced by arrowroot fibre is expected to produce better outcomes. Meanwhile, researchers investigated the impact of blackberry pulp incorporation on the physical, mechanical, and barrier properties of composite films made from arrowroot starch and blackberry pulp (Nogueira, Soares, et al., 2019). Likewise, Fakhouri et al. (2019) investigated the effect of cranberry powder incorporation on the microstructure and thermal properties of arrowroot starch/cranberry powder composites. Recently, De Sá et al. (2015) examined the characterization of nano whiskers cellulose derived from arrowroot fibre.

Up to date, there has been insufficient research on the applications of arrowroot biopolymers, fibres, and biocomposite. There has been a significant amount of work published on arrowroot starch in terms of characterization and application. No research has been conducted on the physicochemical, thermal, and morphological properties of arrowroot fibres. As a result, the focus of this research is on isolating arrowroot fibres and starch from arrowroot plant parts such as tubers and developing arrowroot starch biopolymer composites with arrowroot starch as a matrix.

1.2 Problem statements

Factors such as increased environmental awareness, societal concerns, governmental policies, and the depletion of petrochemical resources have accelerated the rapid growth of new green materials such as eco-friendly packaging films. The proper disposal of petroleum-based plastics, once they have served their purpose, has become a major global environmental issue that must be addressed immediately. Landfilling and incineration are the two most widely used disposal methods. However, due to the significant trash disposal, landfill capacity is rapidly diminishing. Increased pollution from the use of plentiful plastics and pollutants from cremation contribute significantly to environmental health issues. As a result, government laws on using non-renewable and non-biodegradable products have become stricter over time, in order to preserve a clean environment for future generations.

In recent years, most polymers used in packaging are still petroleum-based. The widespread use of petroleum-based materials increases CO₂ emissions, which leads to global climate change. Hence, to reduce reliance on petroleum-based polymers, this research attempted to use a 100% renewable and biodegradable biopolymer derived from arrowroot rhizomes (*Maranta arundinacea* L.). The rhizome of the arrowroot plant is a good source of starch, bagasse fibre, and husk fibre. However, because such a bio-sourced is presently underutilized, very little research on its development as a green packaging material have been reported. As a result, in the current work, arrowroot starch and arrowroot fibres were used to generate completely biodegradable films and biocomposite films as an environmentally friendly food packaging material. However, because of its hydroxyl or polar groups, arrowroot starch, like most other biopolymers,

is hydrophilic. The problems relating to brittleness, processability, high moisture sensitivity, quick retrogradation, poor mechanical and barrier properties, and poor mechanical and barrier characteristics are the major challenges for the development of starches as packaging films (Sanyang et al., 2016a). The limitations can be solved by reinforcing arrowroot starch with arrowroot fibres into high-performance thermoplastic starch for packaging applications. To the best of author's knowledge, no research has been conducted on the characterization of arrowroot fibres and their application in reinforcing biopolymer composite films.

1.3 Research objectives

The overall objective of this research is to develop and characterize environmentally friendly arrowroot (*Maranta arundinacea* L.) fibre reinforced arrowroot starch biocomposite films. The research objectives can be specified into:

1. To characterize the arrowroot fibres from arrowroot tubers (Bagasse, and Husk) to explore their potential to develop a new fully biodegradable and environmentally friendly composite film.
2. To evaluate the effect of glycerol concentration on the physical, mechanical, thermal, and barrier properties of arrowroot starch-based films.
3. To determine the physical, morphological, and mechanical properties of arrowroot fibre reinforced arrowroot starch biocomposites at different fibre loading.
4. To determine the thermal, biodegradability, water barrier, and antimicrobial properties of arrowroot fibre reinforced arrowroot starch biocomposites at different fibre loading.

1.4 Significance of the research

1. This research aims to contribute to this growing area of study by investigating the information in developing high-performance biodegradable bio-packaging films derived from arrowroot starch.
2. To address environmental problems, which arise from the nonbiodegradable disposal, such type of environmentally friendly packaging films (100% biodegradable polymer composites) which is made by a single source (arrowroot) for starch and fibre can be used (instead of the use of synthetic polymer and synthetic polymer composites).
3. This research also reduced the wastage issue by providing the platform for making use of agricultural products wastage into fibres and biopolymers.

4. The successful development of such green materials from arrowroot would provide good opportunities to improve the standard of living of the farmers who cultivated arrowroot by developing economically in rural areas.
5. This research may also include the effort to reveal the potential of arrowroot starch and fibre in developing green products; else, such plentiful resources may be underutilized.

1.5 Scope of research

This research focused on the extraction of biopolymers and natural fibres from arrowroot tubers, as well as the production of biocomposites. A series of experiments were conducted to characterize arrowroot starch and fibres, as well as biopolymers and biocomposite films. The films were developed using a solution casting method with the addition of glycerol. The biocomposite films were developed in three stages using arrowroot fibres and arrowroot starch. The first phase involved investigating the physicochemical, morphological, and thermal properties of arrowroot starch and fibres in order to determine the feasibility of using arrowroot starch and fibres to develop materials with good thermal and mechanical properties for packaging applications. The second phase involved investigating the effect of varying concentrations of glycerol plasticizer on the physical, thermal, morphological, biodegradability, and mechanical properties of starch-based films. In the final phase, the characterized arrowroot fibre was utilized as reinforcement for biopolymer films to improve the matrix properties. Thus, the effect of arrowroot fibre loading (0-10 wt.%) on the physical, thermal, mechanical, water barrier, and antimicrobial properties of biocomposite films was investigated.

1.6 Structure of the thesis

The thesis structure follows Universiti Putra Malaysia alternative thesis format based on publications. Each research chapter (4–7) signifies distinct research that has its own: ‘Introduction’, ‘Materials and methods’, ‘Results and discussion’, and ‘Conclusion’. Further details on the thesis structure are as presented below.

Chapter 1

This chapter clearly describes the problem statement and research objectives. This chapter also demonstrated the significance and contribution of the research, as well as the scope and limitations of the research.

Chapter 2

This chapter provides a comprehensive review of the literature in the key areas related to the title of this thesis. In addition, the chapter discusses the research gaps identified through the literature review.

Chapter 3

This methodology chapter contains every activity related to this research, from material preparation to material processing, testing methods, and data collection and analysis.

Chapter 4

This chapter presents the first article entitled “Extraction and Characterization of a Novel Natural Lignocellulosic (Bagasse and Husk) Fibres from Arrowroot (*Maranta arundinacea* L.)”. In this article, the physical, morphological, structural, and thermal properties of arrowroot bagasse and husk fibres were investigated.

Chapter 5

This chapter presents the second article entitled “Effect of glycerol plasticizer loading on the physical, mechanical, thermal, and barrier properties of arrowroot (*Maranta arundinacea* L.) starch biopolymers”. In this article, the effect of glycerol plasticizer at different concentrations (15, 30, and 45%) on the properties of the arrowroot starch-based film was investigated.

Chapter 6

This chapter presents the third article entitled “Physical, mechanical and morphological performances of arrowroot (*Maranta arundinacea* L.) fibre reinforced arrowroot starch biopolymer composites”. The aim of this article was to develop and characterize biocomposite films based on arrowroot starch matrix and arrowroot fibre as reinforcement filler at different loadings.

Chapter 7

This chapter presents the fourth article entitled “Thermal, flammability, and antimicrobial properties of arrowroot (*Maranta arundinacea* L.) fibre reinforced arrowroot starch biopolymer composites for food packaging applications”. This article studied the effect of various loading of arrowroot fibre (2%, 4%, 6%, 8%, and 10%) on the thermal, antimicrobial, biodegradability, flammability, and water barrier properties of thermoplastic arrowroot starch-based biocomposite films.

Chapter 8

This chapter presents general conclusions collected from different research articles, as well as relevant suggestions and recommendations for further research.

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