UNIVERSITI TEKNIKAL MALAYSIA MELAKA

INNOVATIVE THERMOPLASTIC STARCH BASED BIODEGRADABLE PACKAGING FOR FOOD INDUSTRIES APPLICATION

This report submitted in accordance with requirement of the Universiti Teknikal Malaysia Melaka (UTeM) for the Bachelor Degree of Manufacturing Engineering (Engineering Materials)

by

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APPROVAL

This report is submitted to the Faculty of Manufacturing Engineering of UTeM as a partial fulfillment of the requirements for the Degree in Bachelor of Manufacturing Engineering (Engineering Materials). The member of the supervisory committee is as follow:

………………………………

Supervisor

ABSTRAK

Penyelidikan ini menunjukkan prestasi komposit plastik biolupus yang menggunakan tepung ubi kayu (TS) dan *polypropylene* (PP) sebagai bahan utama matriks dengan penambahan serat gentian tebu (SCFC) sebagai penguat dan gliserol sebagai bahan tambah pemplastik. Tujuan utama penyelidikan ini adalah untuk membuat bungkusan plastik biolupus untuk industri makanan dengan menggunakan kaedah pencampur dalaman yang dinamakan sebagai *PTSG Biodegradable Hybrid Composite*. Kesan penggunaan hampas tebu di dalam komposit ini dikaji dengan lebih lanjut dan hasil komposit yang baik telah dicadangkan. Sampel komposit dihasilkan dengan menggunakan formula pencampuran yang berbeza dengan menggunakan kaedah campuran dan mampatan. Bagi mencapai objektif penyelidikan ini, pengujianan lanjutan untuk mengenalpasti sifat-sifat komposit tersebut dilakukan melalui ujian tegangan, ujian hentaman, dan ujian lenturan. Ujian-ujian ini dilakukan bagi menentukan sifat-sifat mekanikal bahan komposit tersebut. Kemudian, diikuti dengan penilaian sifat persekitaran *PTSG Biodegradable Hybrid Composite* seperti ujian biolupus, uji penyerapan air dan ujian pembengkakan tebal. Dalam rangka untuk mengetahui morfologi patahan sampel, pemerhatian melalui mikroskop imbasan elektron (SEM) telah dijalankan. Peningkatan kadar tepung ubi telah meningkatkan kadar pelupusan, penyerapan air dan pembengkakan tebal yang dihasilkan oleh komposit. Perumusan yang terbaik adalah pada kombinasi 90% *PP*, 10% *TS* 1% *SCFC* dan 3% *glycerol* untuk *PTSG Biodegradable Hybrid Composite* yang mempunyai potensi kinetik pelupusan sebanyak 0.014%/hari bagi penurunan berat yang memberikan cirri-ciri potensi dalam industri pembungkusan makanan. Secara keseluruhan, semua tujuan kajian yang dinyatakan telah tercapai dengan jayanya.

ABSTRACT

This research shows the performance of biodegradable plastic composite using tapioca starch (TS) and polypropylene (PP) as a primary matrix material with addition of sugar cane fiber cellulose (SCFC) as reinforcement and glycerol as additive plasticizer. The main objective of this research is to fabricate a biodegradable plastic packaging for food industries using an internal mixer method which produced the novel biocomposites that simply named as PTSG Biodegradable Hybrid Composite. The effect of SCFC loading was studied as to establish the best formulation of the PTSG Biodegradable Hybrid Composite. The composites were fabricated into thin sheets using a hot compression molding machine. The PTSG Biodegradable Hybrid Composite were tested for tensile test, impact test, and flexural test. Then, the environmental properties were determined through soil burying test, weathering test, water absorption test and thickness swelling test. The fracture morphology of the samples was observed under scanning electron microscope (SEM). The increasing of tapioca starch content had significantly increased the degradation, water absorption and swelling thickness of produced composite. The optimum compounding formulation of the fabricated composites was observed at the combination of 90 wt.% of PP, 10 wt.% of TS, 1 wt.% of SCFC and 3 wt.% of glycerol for PTSG Biodegradable Hybrid Composites that have the potential kinetic of degradation at 0.014 %/day of weight reduction which could potentially applied in the real green food packaging industries as one of new candidates for future food packaging materials. In overall, all the listed research objectives were successfully achieved from this preliminary research

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DEDICATION

Emak, Abah and All My Family Members

Along, Angah, Kak De, Kak Nak, Atam, Atih, Achik, Mamat, Ida

My Bear

My Lecturer, Che Jep

Lovely Friends 4 BMFB 2011

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LIST OF ABBREVIATIONS

ASTM	American Standard Testing of Materials
CMC	Ceramic Matrix Composite
DS	Degrees of Substitution
DSC	Differential Scanning Calorimetry
Eg.	Example
et al.	and others
etc.	Et cetera
LDPE	Low Density Polyethylene
LLDPE	Linear Low Density Polyethylene
MAPP	Maleic-Anhydride Grafted Polypropylene
MMC	Metal Matrix Composite
MS	Modified starch
PC	Polycarbonates
PCL	Polycaprolactrone
PHA	Polyhydroxyalkanoate
PHBV	polyhydroxybutyrate and hydroxyvalerate
PLA	polylactic acid
PMC	Polymer Matrix Composite
PP	Polypropylene
TS / SCFC	Tapioca starch reinforced sugar cane fiber cellulose
S	Starch
SCFC	Sugar Cane Fiber Cellulose
SEM	Scanning electron microscopy
S-g-OSA	Succinylated Starches
TGA	Thermo Gravimetric Analyzer
TS	Tapioca Starch
Wt%	Percent of weight fraction
WA	Water Absorption

CHAPTER 1

INTRODUCTION

1.1 Introduction

A plastic material is one of a wide range of synthetic or semi-synthetic organic amorphous solid used in the manufacture of industrial products. Due to their versatility, plastic is great for packaging of a variety products such as processed and convenience foods, pharmaceuticals and medicines, cosmetics and toiletries, household and agricultural chemicals, petroleum product and detergent. Nowdays, plastic containers are successfully replacing glass, tin, metal, aluminum and paper containers in many applications. This is because plastics are typically polymers of high molecular mass, and may contain other substances to improve performance of the fabricated products. Therefore, the advantages of plastics are light and less bulky than other packaging materials that could be processed into any desired shape or form such as films, sheets and pouches. On the other hand, it saves costs of storage and transportation because of lower volume, easy coloring, no rusting and first-rate water resistance. Although plastic package have great advantages, however they also have some limitations that includes some chemical attack on particular plastics, tendency to creep, less heat resistance, lower gas barrier and lower dimensional stability (Athalye, 2002).

Into the bargain, there are grave problems connected with the analytical control of such materials which are toxic hazards from the modified plastics and also from their degradation products, increased costs and the possible encouragement of litter. In order to trim down this problem, the application of biodegradable material as an alternative choice is increasingly applied. Biodegradable which are often produced from renewable sources, are being increasingly sought after by food processors as part of a solution to environmental concerns over waste and the use of fossil fuels. The process is called biodegradation (Dong *et al.* 2008). Biodegradation is a natural process by which organic chemicals in the environment are converted to simpler compounds, mineralized, and redistributed through the elemental cycles such as the carbon, nitrogen, and sulphur by the action of microorganism.

In this research, biodegradable polymer matrix composites (PMCs) were developed. There are two natural components were combined which are tapioca starch (TS) as a natural matrix material and sugar cane fiber cellulose (SCFC) as a natural biofiber. Sugar cane has good performance as filler reinforcement, together with the advantages to the environment when it combines with the tapioca starch matrix. Polypropylene (PP) and glycerol are being utilized as part of two mixtures as to produce the innovative formulation as well as to facilitate the processing. It is expected that the development of this product, will contribute to the world as novel biodegradable, non-toxic and non-allergenic bio environmental friendly natural green products.

There is considerable interest and noble aims in this research where to produce an alternative material by compounding tapioca starch (TS), polypropylene (PP), sugar cane fiber cellulose (SCFC) and glycerol to replace the existing non biodegradable plastic material in the commercial market. Thus, overall of this research is to formulate the biodegradable based composites filled with an agro-waste biofiller by using the internal mixer method in order to investigate and understand the behavior and properties as well as characteristic of the tapioca starch (TS), polypropylene (PP), sugar cane fiber cellulose (SCFC) and glycerol biocomposites or in short is namely as PTSG Biodegradable Hybrid Composite. Utilization of biodegradable based product will significantly reduce the need for synthetic polymer production at low cost, thereby producing a positive effect both environmentally and economically.

1.2 Objective

The purposes of this study are:

- To formulate biodegradable packaging material for food industries by using internal mixer method combining the tapioca starch (TS), polypropylene (PP), sugar cane fiber cellulose (SCFC) and glycerol, in their formulations.

- To establish and understand the mechanical, morphological and degradation behavior of novel biocomposites containing tapioca starch (TS), polypropylene (PP), and sugar cane fiber cellulose (SCFC) and glycerol mixtures, in comparisons to other biocomposites.

1.3 Problem Statement

There are problems connected with the utilization of hydrocarbon plastic based materials which are toxic hazards from the modified plastics and also from their degradation products, high costs and possible encouragement of litter. Allocated to this problem, it is important to produce innovative products which combine the utilization of natural filler with other elements such as glass, plastics and synthetics material. Ecological concerns have resulted in overwhelming interest in natural and compostable materials and issues related with the biodegrability and environmental safely that are becoming corresponding important (Yu *et al*., 2007). Biodegradable or environmentally acceptable materials have attained increasing interest in few decades to the researchers and manufactures due to environmental pressure derived from the consumption of petroleum based materials, difficulties in degradation in a landfill and composting environments (Liu *et al*. 2006; Huda *et al*. 2007). Starch is one of the most promising materials for biodegradable plastics because of its versatile biopolymer characteristic with immense potential and low price for application in the food industries (Mohanty *et al*., 2000). However, starch was mostly water soluble, difficult to process and brittle in nature when it is used without the addition of plasticizer. Furthermore, the mechanical properties are very sensitive to moisture content, which is difficult to control and predict. In principle, some properties of starch could be significantly improved by blending it with polymer (Dufresne *et al.*, 2000). As a result, the TS, PP, SCFC and glycerol are combined as to formulate the new potential biodegradable packaging material for food industries application.

1.4 Significant of Study

In this research, the noble aim is to develop the PTSG Biodegradable Hybrid Composite for the food packaging application. As a consequence, by conducting this research, it is expected that it will be benefited to the environment that suffer with non-degradable waste of plastic food packaging caused by uncontrolled solid waste disposal and extensive utilization of this necessity. Development of this novel food packaging alternative will create potential solution to the environmental friendly and safe packaging medium either for food, consumer or environment as a whole.

1.5 Thesis Overview

This thesis is divided into five chapters that describe the analytical and experimental research performed. The First Chapter is an introduction to the study that brief about objectives, problem statement, significant of study and the thesis overview. Chapter Two present the literature review that relates to the theories on composites and previous investigations on the biocomposites. The important element that included in this chapter is about the types of properties for SCFC and TS and also related experimental testing. Chapter Three, provides details explanations on the methodology used for overall research work, raw materials, procedure property analysis that had been done. In Chapter Four, the result of the characterization and measurement of various engineering properties for the fabricated biocomposites were explained in details. The final chapter (Chapter 5) concludes the overall results obtained from this research. In this chapter, it explains either the objectives of this study are achieved or not. The recommendation for future project also has been included in this Chapter 5.

CHAPTER 2
LITERATURE REVIEW

2.1 Introduction

This chapter review related studies done by previous researcher on the tapioca starch (TS) as matrix whiles the sugar cane fiber cellulose (SCFC) as reinforcement in the biocomposites product based development. Through this chapter, the study on the SCFC, TS and Polypropylene (PP) engineering properties had been highlighted and extensively reviewed. The literature review mainly focused on the various types of engineering properties and related experimental testing will be studied and discussed. The fundamental of composites were reviewed in the first following section and followed by the engineering properties from the existing research of the biocomposites, as well as the degradation of the composites.

2.2 Composites

A composite material is a material system that composed of a suitably arranged mixture or combination of two or more micro or macroconstituents with an interface separating them that differ in form and chemical composition and are essentially insoluble in each other (Smith *et al*. 2000). An engineering importance of a composite material is that two or more distinctly different materials combine to form a composite material that possesses properties that are superior, or important in some other manner, to the properties of the individual components (Smith *et al*. 2000).

Thus, composites are those materials formed by aligning extremely strong and stiff constituents such as fibers and particulates in a binder called matrix. The materials in this category have excellent mechanical properties. There are two components in the composite, which are reinforcing phase and matrix phase. Polymers, ceramic and metals have found their application as matrix materials. The reinforcing phase is other component and is called reinforcement and could be fiber, particulate or laminar (Gupta, 2005). According to Matthews and Rawlings (2002), each of materials (i.e., matrix and reinforcement) must exist of more than 5 wt% to be classified as composites material. The purposed of introducing the composites material is to improve the properties that have been performed by monolithic material. Thus, the composite is expected to improve the mechanical characteristics such as stiffness, toughness, ambient and high-temperature resistance (Callister, 2003).

According to Callister (2003), the composites materials are classified by the geometry of reinforcement and the types of matrix materials used in its constituent. As a result, there are three main types of geometries of the reinforcement which are particle-reinforced, fiber-reinforced and structural composites (Callister, 2003). Types of matrix used in composites also can be divided into four types, namely as polymer-matrix composite (PMC), metal-matrix composites (MMC), ceramics-matrix composites (CMC), and carbon-carbon composites (CCC) (Matthews and Rawlings, 2002; Callister, 2003; Taj *et al*. 2007). In this study, PMC becomes as the topic of investigation.

2.3 Polymer Matrix Composites (PMC)

As the terms implied, the polymer matrix composites consists of polymeric material as the matrix material and fiber as the reinforcement elements (Luz *et al*. 2007). Research and engineering interest has been shifted from the monolithic material to fiber-reinforced polymeric material (Wambua *et al*. 2003). Polymer matrix composite is the common types of matrix composites used for a few decades. The enormous applications of PMC have exhibited various performances that cannot be performed by any monolithic material (Wambua *et al*. 2003).

There are three types of major classes in polymeric materials which are thermoplastics, thermosets, and elastomers (Kalpakjian, 2006). Reinforcement of polymers by strong fibrous network permits fabrication of PMC characterized by the properties such as high tensile strength, high stiffness, high fracture toughness, good abrasion resistance, good corrosion and puncture resistance but the main disadvantages are low thermal resistance and high coefficient of thermal expansion (Liu *et al*. 2007; Taj *et al*. 2007).

The extensive used of PMC as daily life components, exhibits that the PMC has low density, ρ where the benefit of low density becomes apparent when the tensile modulus per unit mass, E/ρ (specific modulus), and tensile strength per unit mass, σ/ρ (specific strength), are considered. Contrarily, it is also observed that the used of PMC has resulted in the reduction of flexural and tensile strength of the fabricated composites (Wambua *et al*. 2003).

There were limitations of PMC which need to be considered, such as their low maximum working temperatures, high coefficient of thermal expansion, dimensional instability, and sensitivity to radiation and moisture (Hanlon *et al.* 1998; Selke *et al.* 2004; Harper, 2006). This leads to a degree of environmental degradation greater than that experienced by the component of material alone. Until early 1970s, the focus was given on the preventing the plastic degradation to avoid the loss in the performance of the plastic properties. The extent of degradation was generally measured by the loss percentages of the useful properties. It was stated that 90 percent loss in the tensile strength was equivalent to total degradation, as this was sufficient to render the plastic object unusable. In the middle of 1980s, when concerns about solid waste disposal were increased, the interest in biodegradation intensified as some perceived it as a solution to the landfill crisis (Hanlon *et al.* 1998; Selke *et al.* 2004; Harper, 2006).