MECHANICAL PROPERTIES AND SURFACE QUALITY OF NATURAL RUBBER-POLY (LACTIC ACID) COMPOSITES FOR FUSED DEPOSITION MODELLING TECHNOLOGY

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

by

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APPROVAL

This report is submitted to the Faculty of Manufacturing Engineering of Universiti Teknikal Malaysia Melaka as a partial fulfillment of the requirement for a Degree of Manufacturing Engineering (Hons.). The members of the supervisory committee are as follow:

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(DR RAHIMAH BINTI ABDUL HAMID)

ABSTRAK

Teknologi pembuatan tambahan membina bahagian lapisan demi lapisan melalui pemendapan bahan tercair mengikut reka bentuk 3D digital. FDM telah menjadi yang paling lazim dalam teknologi percetakan 3D. Poli (asid laktik) atau PLA, adalah polimer termoplastik dalam percetakan 3D yang boleh terbiodegradasi dan berpotensi untuk aplikasi bioperubatan. Kelemahan PLA ialah rapuh, mempunyai keliatan yang rendah, dan tidak fleksibel dalam bentuk tulennya. Disebabkan ini, banyak usaha telah dijalankan untuk membangunkan bahan tambahan yang berkesan untuk pengubahsuaian PLA bagi memperluaskan aplikasi. Oleh itu, nisbah campuran polimer dan bahan tambahan yang betul perlu dikenal pasti. Dalam kajian ini, keserasian campuran berasaskan NR dan PLA untuk fabrikasi filamen dalam teknologi FDM melalui analisis sifat mekanikal. Pada mulanya, adunan telah disediakan mengikut nisbah NR/PLA 10/90, 20/80, 30/70, 40/60, 50/50, 60/40, 70/30, dan 90/10, dan mencairkan sebatian dengan pengadun dalaman. Selepas itu, sifat mekanikal spesimen PLA diperkuatkan NR yang dibuat melalui mesin tekan panas dibandingkan dengan PLA tulen untuk kekuatan tegangan dan lentur. Kompaun spesimen fabrikasi telah dibentuk menggunakan kaedah mampatan tekan panas yang mematuhi ASTM-D638 (Jenis IV) dan ASTM-D790, diuji menggunakan Mesin Pengujian Universal Shimadzu (UTM) pada kelajuan 5 mm/minit dan 50 mm/minit. Profilometer Mitutoyo Surftest SJ-301 digunakan untuk memeriksa kualiti permukaan (R_a) spesimen PLA diperkuatkan NR berbanding spesimen PLA tulen yang direka melalui cetakan 3D. Keratan rentas spesimen tegangan patah digunakan untuk analisis struktur mikro untuk mengkaji mekanisme pengadunan NR/PLA. Keputusan kajian ini menunjukkan bahawa komposisi 40 phr NR/PLA meningkatkan kekuatan tegangan dan lentur berbanding dengan PLA tulen. Walau bagaimanapun, 20 phr NR/PLA merupakan nilai R_a yang lebih baik untuk spesimen yang ditekan panas. Selain itu, imej SEM menunjukkan bahawa saiz titisan NR meningkat dengan penurunan kandungan getah. Kesimpulannya, nisbah adunan NR40/PLA60 didapati terbaik untuk fabrikasi filamen FDM memandangkan sifat mekanikal lebih memudaratkan daripada kekasaran permukaan dalam pertimbangan ini. Sebagai cadangan, analisis lanjut bagi komposit NR/PLA hendaklah dijalankan untuk DSC, FTIR, ujian kekerasan, ujian hentaman, ujian penyerapan air, atau XRD untuk menganalisis lebih lanjut sifat komposit NR/PLA ini. Ini penting dengan mengambil kira kos pelet PLA, sebagai tambahan kepada masa yang diperlukan untuk penyediaan jumlah adunan NR/PLA yang mencukupi sebelum filamen NR/PLA boleh dihasilkan melalui penyemperit skru tunggal.

ABSTRACT

Additive Manufacturing (AM) technology builds parts layer by layer through the deposition of liquidized material according to digital 3D design data. Fused Deposition Modelling (FDM) has been the most prevalent in 3D printing technology. Poly (lactic acid) or PLA, is one of the thermoplastic polymers in 3D printing that is biodegradable and has potential for biomedical applications. The only drawback is that the PLA is brittle, has low toughness, and inflexibility in its pure form. Due to this, many efforts have been undertaken to develop effective additives for PLA modification in order to expand application possibilities. Therefore, the correct blending ratio of the polymer and additives needs to be identified. In this study, the compatibility of NR and PLA-based blends were investigated for filament fabrication in FDM technology through mechanical properties analysis. Initially, the blends were prepared according to NR/PLA ratios of 10/90, 20/80, 30/70, 40/60, 50/50, 60/40, 70/30, and 90/10, and melt compounding with an internal mixer. Subsequently, the mechanical properties of NR-reinforced PLA specimens fabricated via a hot-pressed machine were compared with the pure PLA for tensile and flexural strength. The compound of fabricated specimens was moulded using a hot-press compression method that conforms with the ASTM-D638 (Type IV) and ASTM-D790, tested using the Shimadzu Universal Testing Machine (UTM) at the speed of 5 mm/minute and 50 mm/minute, respectively. A Mitutoyo Surftest SJ-301 profilometer was used to examine the surface quality (R_a) of the NR-reinforced PLA specimens compared to the pure PLA specimen fabricated via 3D printing. The cross-sectional of the fractured tensile specimen was used for microstructure analysis to study the blending mechanism of the NR/PLA. The result of this study shows that the 40 phr NR/PLA composition enhances the tensile and flexural strength compared to the pure PLA. However, the 20 phr NR/PLA offers a better R_a value for the hot-pressed specimen. Other than that, the SEM images indicated that the size of NR droplets increases with decreasing rubber content. In conclusion, a blending ratio of NR40/PLA60 was found to be the best for FDM filament fabrication in future work since the mechanical properties are more detrimental than the surface roughness in this judgement. As a recommendation, a further analysis of the NR/PLA composites shall be conducted for DSC, FTIR, hardness test, impact test, water absorption test, or XRD to further analyze the properties of these NR/PLA composites. This is important with consideration of the cost of PLA pellets, in addition to the time needed for the preparation of a sufficient amount of NR/PLA blends before NR/PLA filament could be manufactured via a single screw extruder.

DEDICATION

Only

my beloved father, Alias bin Ismail

my true loved mother, Halijah binti Sharip

my adored brother, Muhammad Asyraf bin Alias

for giving me moral support, money, cooperation, encouragement, and also understanding

Thank You So Much and I Love You All Forever

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LIST OF ABBREVIATIONS

2D	-	Two Dimensional
3D	-	Three-Dimensional
ABS	-	Acrylonitrile Butadiene Styrene
AC	-	Alternating Current
AM	-	Additive Manufacturing
ASTM	-	American Society for Testing and Materials
Au	-	Gold
CAD	-	Computer Aided Design
CNT	-	Carbon Nanotube
CSS	-	Core Shell Shape
DRC	-	Dry Rubber Content
EGMA	-	Ethylene Glycidyl Methacrylate
ENR	-	Epoxidized Natural Rubber
FDM	-	Fused Deposition Modelling
FFF	-	Fused Filament Fabrication
FKP	-	Fakulti Kejuruteraan Pembuatan
GMA	-	Glycidyl Methacrylate
GPTMS	-	Glycidyloxypropyl Trimethoxysilane
HH	-	Hemp Hurd
ISO	-	International Organization for Standardization
LCD	-	Liquid Crystal Display
MCC	-	Microcrystalline Cellulose
NaOH	-	Sodium Hydroxide
NR	-	Natural Rubber
PA	-	Polyamide
PBAT	-	Polybutylene Adipate Terephthalate
PBF	-	Powder Bed Fusion
PC	-	Polycarbonate
Pd	-	Palladium

PE - Polyethylene
PEEK - Polyether Ether Ketone
PEI - Polyetherimide
PET - Polyethylene Terephthalate
PETG - Polyethylene Terephthalate Glycol
PLA - Polylactic Acid
PP - Polypropylene
PPSF - Polyphenylsulfone
PSM - Projek Sarjana Muda
RP - Rapid Prototyping
SEM - Scanning Electron Microscope
STL - Stereolithography
TBC - Tributyl Citrate
Tg - Glass Transition Temperature
TPU - Thermoplastic Polyurethane
UTeM - Universiti Teknikal Malaysia Melaka
UTS - Ultimate Tensile Strength
VPP - Vat Photopolymerization

LIST OF SYMBOLS

α - Alpha
°C - Degree Celsius
° - Degree
% - Percent
g - Gram
g/cm^3 - Gram per Centimetre Cubic
g/min - Gram per Minute
g/ml - Gram per Millilitre
g/mol - Gram per Molecule
GPa - Gigapascal
K - Thousand
kg - Kilogram
kN - Kilo Newton
mm - Millimetre
MJ/m^3 - Megajoules per Metre Cubic
ML - Mooney Viscosity
mm/min - Millimetre per Minute
mm/s - Millimetre per Second
mol% - Mole Percent
MPa - Megapascal
N - Newton
N/mm^2 - Newton per Square Millimetre
nm - Nanometre
phr - Part per Hundred of Rubber
Ra - Average Roughness
rpm - Revolution per Minute
wt% - Weight Percentage
Zi - Standard Deviation
µm - Micrometre

CHAPTER 1

INTRODUCTION

This chapter describes the preface of this investigation work which includes the background, problem statement, objectives, scope, and significance of the study. The experimentation of the influence of adding Natural Rubber (NR) as fillers with Polylactic acid (PLA) composites on the mechanical properties and surface quality is carried out in this work.

1.1 Background of Study

Additive Manufacturing (AM) or 3D printing technology has developed importance in every field due to its affordability and simple working principle for creating complex geometries from 3D CAD data. The technology enables the manufacture of customized products without requiring manufacturing costs as no special tools or moulds are needed. Due to the accessibility of relatively low cost 3D printing equipment, the use of AM technology has increased dramatically in both quantity and range. More than that, AM technology makes the rapid production of complex and integrated functional designs possible that potentially reducing the exigency of assembly work.

Fused Deposition Modelling (FDM) is a well known technique for 3D printing technology with a relatively low cost, quick, and easy printing process for a wide range of thermoplastic polymer materials. The low cost FDM 3D printer is affordable but also portable and simple to maintain compared to other AM technologies. FDM allows the production of solid parts made of thermoplastic polymers which extrude a material onto the build platform according to the desired CAD design. The thermoplastic polymers form a filament that is heated and transformed to a semi-liquid state and injected into a nozzle following the input from the slicing software.

Thermoplastic polymers like polylactic acid (PLA), polyethylene (PE), polypropylene (PP), and acrylonitrile butadiene styrene (ABS) are some types of materials used in FDM. Park and Fu (2021) claimed that PLA is an amorphous, durable, strong thermoplastic material that is healthy, pollution free, and prints exceptional. As a result, PLA is a frequently used material in 3D printing. Torres *et al.* (2016) supported that PLA is amongst the most widely recognized plastics for 3D printing and is economically beneficial due to its low melting temperature and outstanding biodegradability. PLA is a common thermoplastic polymer material in FDM with a broad assortment of potential applications due to its biodegradability and biocompatible characteristics and mainly used (Pakkanen *et al.*, 2017).

Natural Rubber (NR) may be used as a filler for a matrix material in natural polymers. NR is derived from the rubber tree, scientifically known as *Hevea Brasiliensis*. NR is biodegradable, inexpensive, and poses no health risks. In other respects, NR is a highly versatile and widely used elastomer because of its abrasion resistance, durability, elasticity, flexibility, and excellent strength. Due to this fact, the combination of PLA and NR as the filler might have many possible applications, particularly in the biomedical field. NR mechanical properties can be improved by adding fillers to the NR which can act as reinforcements such as carbon black (Karasek and Sumita, 1996) and silica (Zeng *et al.*, 2009). There has been an increasing interest in enhancing the performance of composites through renewable reinforcements. Composite materials can be created by meticulously combining and controlling the properties of various materials. The effect of a particular filler differs depending on the polymer. Skelhorn (2003) confirmed that one way to tell those apart is through their impact on pure natural rubber. A reinforcing filler can improve abrasion resistance with tensile and tear strengths. The softening of plastic strain is characterized by matrix localization caused by multiple crazing, extensive yielding under shear, or a combination of these elements.

Enhancing the polymer toughness is critical for a wide variety of interrelated applications. Toughness and fracture toughness are terms used to refer to the physical absorption of energy during the emergence of a deformation in a fracture. Typically, polymer modification aims to create a material that possesses a significant plastic elongation at break while retaining desirable properties of comparatively stiffness and strength augmented by a dominant high toughness. This can be done by altering the structure in a particular way that facilitates plastic deformation capable of absorbing energy. One of the most significant disadvantages of polymers is their low resistance to impact. In order to further enhance this performance, considerable effort has been spent incorporating an elastomeric phase dispersed within a polymer matrix. Currently, the rubber toughening in polymer performance is undergoing extensive research. Be that as it may, it is discerning to establish that the toughened polymer can become brittle because temperatures are relatively low or have high loading rates. Some suggest that the brittleness of toughened polymers is caused by the glass transition in the rubbery phase at low temperatures. Therefore, this study aims to evaluate the NR/PLA performance at different NR/PLA compositions to identify the best blending compositions for the NR/PLA FDM filaments as the potential application. The emphasis is given to the mechanical properties behavior, which is critical for biomedical applications.

1.2 Problem Statements

FDM is one of the most popular AM technologies due to its affordability and simple technology (Masood and Song 2004). The thermoplastic polymer is used which includes ABS, PP, PLA, TPU, PETG, PEEK, and PA (PA6, PA12). However, FDM technology's manufacturing process involves the extrusion of pre-heated polymers and offers lower mechanical properties, strength, and toughness (Oksiuta *et al.*, 2020). Rezvani Ghomi *et al.* (2021) found that PLA has a biodegradability property from natural sources. However, PLA is brittle while having a low toughness and flexibility in a pure form (Farah *et al.*, 2016). Due to these limitations, its prospective applications that need excellent impact toughness and flexibility are limited. Various attempts have been made to identify viable additives for PLA modifications in order to enable enhanced applications.

According to Deb and Jefferson (2021), PLA has a low deformation at break and a low resistance to impact. PLA is typically modified to overcome these drawbacks by