

PHYSICAL, MECHANICAL AND MORPHOLOGICAL PROPERTIES OF RECYCLED POLYPROPYLENE REINFORCED WITH SUGAR PALM FIBER FILAMENT



BACHELOR OF MANUFACTURING ENGINEERING TECHNOLOGY WITH HONOURS

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Bachelor of Manufacturing Engineering Technology with Honours

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2022

DECLARATION

I declare that this thesis entitled "Physical, Mechanical and Morphological Properties of Recycled Polypropylene Reinforced With Sugar Palm Fiber Filament" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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APPROVAL

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DEDICATION

This study is specially dedicated to my beloved parents, who have given the strength that I deserved when I thought of giving up, always providing their moral, spiritual, emotional and financial support.

Not to forget my sibling, sister and brother, relatives, academic advisor and supervisor Dr.

Nuzaimah Binti Mustafa, friends and classmates who shared their words of advice and

encouragement to finish this study.

Last but not least, I dedicated this book to The Almighty "Allah", thank you for the guidance, strength, power of mind, protection, skills and for giving a healthy life.

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ABSTRACT

Presently, polypropylene (PP) is plastic-based material that being widely used for commercial and household purposes along with polyethylene terephthalate (PET) and other thermoplastic material. The development of biodegradable polymer from recycling resources has been promoted by increasing environmental awareness to replace conventional non-biodegradable polymer in various applications. PP is one of the most affordable plastics available today, and it is used both as a plastic and a fiber in sectors such as automotive manufacturing, furniture assembly, and the aerospace sector. PP offers several benefits such as low cost, high melting point, sustainable and most important thing is, it is 100% recyclable. However, PP has become a major threat to the environment due to its nondegradable property. There are certain benefits of using natural fibers as reinforcement in 3D printing filament due to the rapid demand for renewable, cost-effective, and eco-friendly materials in many applications. The addition of natural fiber to polymer composites as reinforcement has created more "green" composites and can replaced conventional glass fiber and other synthetic fiber composites that possess manufacturing hazard. As a natural fiber, SPF is the key component that become one of the element for reinforment polymer composites. Sugar palm fiber (SPF) was extracted from sugar palm tree (Arenga Pinnata) which were usually found in Southeast Asia like Malaysia and Indonesia. In this study, SPF was successfully treated by alkaline treatment using 6% of sodium hydroxide solution (NaOH). The investigations of thermal properties were then carried out by using thermogravimetry analysis (TGA) and diffraction scanning calorimetry (DSC). The fabrication of 3D printing filament using twin screw extruder machine by inserting different amount of sugar palm fiber loading (0%, 1%, 3%, 5%) into the polymer matrix as reinforcement to find the characteristic of their physical and environmental properties. A few tests have been done to analyze the physical, mechanical, and morphological properties of the filament. Overall, recycled polypropylene reinforced with sugar palm fiber are potential alternative to develop a new 3D printing filament material.

ABSTRAK

Pada masa ini, polipropilena (PP) adalah bahan berasaskan plastik yang digunakan secara meluas untuk tujuan komersial dan isi rumah bersama dengan polietilena tereftalat (PET) dan bahan termoplastik lain. Pembangunan polimer terbiodegradasi daripada sumber kitar semula telah digalakkan dengan meningkatkan kesedaran terhadap alam sekitar untuk menggantikan polimer tidak terbiodegradasi konvensional dalam pelbagai aplikasi. PP adalah salah satu plastik yang paling berpatutan yang ada sekarang, dan ia digunakan sebagai plastik dan gentian dalam sektor seperti pembuatan automotif, pemasangan perabot, dan sektor aeroangkasa. PP menawarkan beberapa faedah seperti kos rendah, takat lebur yang tinggi, mampan dan yang paling penting ialah, ia 100% dapat dikitar semula. Walau bagaimanapun, PP telah menjadi ancaman besar terhadap alam sekitar kerana harta benda yang tidak boleh terurai. Terdapat faedah tertentu menggunakan gentian semula jadi sebagai penguat dalam filamen percetakan 3D kerana permintaan yang cepat untuk bahan yang boleh diperbaharui, menjimatkan kos, dan mesra alam dalam banyak aplikasi. Penambahan gentian semula jadi untuk komposit polimer sebagai tetulang telah menghasilkan lebih banyak komposit "hijau" dan dapat menggantikan gentian kaca konvensional dan komposit gentian sintetik lain yang mempunyai bahaya pembuatan. Sebagai gentian semula jadi, SPF adalah komponen utama yang menjadi salah satu elemen untuk komposit polimer pengukuhan. Serat kelapa sawit (SPF) diekstrak daripada pokok kelapa sawit (Arenga Pinnata) yang biasanya terdapat di Asia Tenggara seperti Malaysia dan Indonesia. Dalam kajian ini, SPF telah berjaya dirawat dengan rawatan alkali menggunakan 6% larutan natrium hidroksida (NaOH). Penyiasatan sifat terma kemudiannya dijalankan dengan menggunakan analisis termogravimetri (TGA) dan kalorimetri pengimbasan difraksi (DSC). Fabrikasi filamen cetakan 3D menggunakan mesin penyemperit skru berkembar dengan memasukkan jumlah muatan serat kelapa sawit yang berlainan (0%, 1%, 3%, 5%) ke dalam matriks polimer sebagai tetulang untuk mencari ciri sifat fizikal dan persekitaran mereka. Beberapa ujian telah dilakukan untuk menganalisis sifat fizikal, mekanikal dan morfologi filamen. Secara keseluruhan, polipropilena kitar semula yang diperkuat dengan serat kelapa sawit adalah alternatif yang berpotensi untuk menghasilkan bahan filamen percetakan 3D baharu.

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UNIVERSITI TEKNIKAL MALAYSIA MELAKA

TABLE OF CONTENTS

		PAGE
DEC	CLARATION	
APP	ROVAL	
DED	DICATION	
ABS	TRACT	i
ABS	TRAK	ii
ACK	KNOWLEDGEMENTS	iii
ТАВ	BLE OF CONTENTS	iv
LIST	Γ OF TABLES	vii
LIST	Γ OF FIGURES	viii
LIST	COF SYMBOLS AND ABBREVIATIONS	vi
T IST	T OF A DDENDICES	
L1 51	I OF ALLENDICES	ХШ
CHA	INTRODUCTION	14
CHA	APTER 1	14
1.1	Background Study TI TEKNIKAL MALAYSIA MELAKA	14
1.2	Problem Statement	15
1.3	Research Objective	17
1.4	Scope of Research	17
CHA	APTER 2 LITERATURE REVIEW	18
CHA	APTER 2	18
2.1	Introduction	18
2.2	3D printing in general	18
	2.2.1 Commonly used 3D printing technology	19
a a	2.2.2 3D printing filament material	20
2.3	Natural fiber	22
	2.3.1 Types of natural fiber in composite	25 26
24	Natural fiber Reinforced Composite	20 27
<i>∠</i> .⊤	2.4.1 Sugar palm fiber reinforced high impact polystyrene (HIPS)	27
	2.4.2 Jute fiber reinforced polypropylene composite	29
2.5	Polymer as matrix in composite	31
	2.5.1 Type of polymer as matrix in composite	32
	2.5.2 Polypropylene	35

iv

	2.5.3 Recycling of polypropylene	36
2.6	Polypropylene (PP) as matrix in composite	37
	2.6.1 Recycled polypropylene as matrix in composite	38
27	Properties of sugar nalm fiber	38
2.7	2.7.1 Thermal properties of sugar nalm fiber	38
	2.7.1 Physical properties of sugar palm fiber	20 20
28	Properties of recycled polypropylene	
2.0	2.8.1 Thermal properties of recycled polypropylene	42
	2.8.1 Thermai properties of recycled polypropylete	42
2.0	2.8.2 Rheological properties of recycled polypropylene	43
2.9	Summary	45
CHA	PTER 3 METHODOLOGY	46
CHA	PTER 3	46
3.1	Introduction	46
3.2	Raw Material	47
	3.2.1 Sugar Palm Fiber (SPF)	47
	3.2.2 Recycled polypropylene (rPP)	47
	3.2.2 Sodium Hydroxide (NaOH)	48
33	Preparation of material	
5.5	2.2.1 Droporation of sugar palm fiber	40
	2.2.2 Dreparation of recursied polymoryland (rDD)	49 51
2.4	5.5.2 Preparation of recycled polypropytelle (IPP)	51
3.4	Fiber Treatment	52
	3.4.1 Preparation of NaOH solution	52
2.5	3.4.2 Preparation of fiber treatment (SPF)	55
3.5	Characterization of sugar palm fiber (SPF)	54
	3.5.1 Particle size analysis	55
	3.5.2 Thermogravimetric analysis (TGA)	55
3.6	Characterization of recycled polypropylene (rPP) SIA MELAKA	56
	3.6.1 Thermogravimetric Analysis (TGA)	57
	3.6.2 Differential Scanning Calorimetry Analysis (DCS)	57
3.7	Preparation of composite	58
	3.7.1 Mixing of sugar palm fiber and recycled polypropylene	58
	3.7.2 Twin Extruder	58
3.8	Characterization of composite	60
	3.8.1 Physical Properties	60
	3.8.2 Mechanical Properties	61
	3.8.3 Morphological Properties	62
3.9	Summary	63
CHAI	PTER 4 RESULTS AND DISCUSSION	64
СНА	PTFR 4	64
<u>4</u> 1	Introduction	6/
<u>-</u> .1 4 2	Thermal Properties	6/
⊤. ∠	121 TGA and DSC result for recycled Polypropylone	0 4 65
	4.2.2 TGA and DSC result for recycled rotypiopylene 4.2.2 TGA results for treated and untreated SDE	03 47
12	4.2.2 IOA IESUIIS IOI IIEAIEU AIU UIIIIEAIEU SFF	0/
4.3	rnysical properties	68

	4.3.1 Water absorption test	68
4.4	Mechanical Properties	70
	4.4.1 Tensile Test	71
4.5	Morphological Properties	73
	4.5.1 Scanning Electron Microscopy Analysis (SEM)	73
4.6	Summary	75
CHA	APTER 5 CONCLUSION AND RECOMMENDATIONS	76
CHA	APTER 5	76
5.1	Conclusion	76
5.2	Recommendations	77
5.3	Project Potential	78
REF	ERENCES	79
ΔPP	PENDICES	84



UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF TABLES

TABLETITLE	PAGE
Table 2.1 Types of 3D printing.	19
Table 2.2 Advantage and disadvantage of FDM technology (Wang et al., 2017).	20
Table 2.3 Comparison of Natural Polymers and Synthetic Polymers (Shrivastava,	
2018).	34
Table 2.4 Resin identification code based on ASTM D7611 (Khosravani & Reinick	e,
2020).	36
Table 2.5 Residual mass % of pure TPU and TPU-SPF composites (Ausama Al-	
sarraf Mohammed et al., 2016).	39
Table 3.1 Specification of recycled polypropylene.	48
Table 3.2 Specification of Sodium Hydroxide.	49
Table 3.3 Weight ratio of water	53

LIST OF FIGURES

FIGURE TITLE	PAGE	
Figure 2.1 FDM setup (Wang et al., 2017).	20	
Figure 2.2 Classification of natural fibers (Hasan et al., 2020).	22	
Figure 2.3 Sugar palm tree (Arenga pinnata) (Sahari et al., 2013).	23	
Figure 2.4 Sugar palm fiber (S et al., 2020).	24	
Figure 2.5 (a) jute plant, (b) raw jute fiber, (c) jute sticks and (d) woven jute fiber.	25	
Figure 2.6 Applications of polymeric based composites (Pupure et al., 2015).	26	
 Figure 2.7 Tensile strength of SPF-HIPS composites for different fibre loadings (Sapuan & Bachtiar, 2012). Figure 2.8 Tensile modulus of SPF-HIPS composites for different fibre loadings 	28	
(Sapuan & Bachtiar, 2012). Figure 2.9 The fractured surfaces of SPF/HIPS composites after tensile testing for:		
(a) 10%; (b) 20%; (c) 30%; (d) 40% and (e) 50% (Sapuan & Bachtiar,		
2012).	29	
Figure 2.10 Tensile strength (MPa) (Chestee et al., 2017).	30	
Figure 2.11 Elongation at break (%) (Chestee et al., 2017).	30	
Figure 2.12 Tensile modulus (GPa) (Chestee et al., 2017).	30	
Figure 2.13 Bending strength (MPa) (Chestee et al., 2017).	31	
Figure 2.14 Bending modulus (GPa) (Chestee et al., 2017).	31	
Figure 2.15 Bonding of monomers to form a polymer (Shrivastava, 2018).	32	
Figure 2.16 (A) Latex from the rubber tree and (B) Honey is an natural polymer from		
bees (Shrivastava, 2018).	33	

Figure 2.17 The resin identification code for polypropylene (Thomas, 2012).	35
Figure 2.18 TGA result of pure TPU and TPU-SPF composites (A. A. Mohammed et	
al., 2016).	39
Figure 2.19 SEM of (a) pure TPU, (b) SPF, (c) 160 μ m TPU-SPF, (d) 250 μ m TPU-	
SPF, and (e) 425 μ m TPU-SPF (A. A. Mohammed et al., 2016).	41
Figure 2.20 TGA curve of RH fiber, rPP and rPP/RH composite filaments obtained	
using TGA (Morales et al., 2021).	43
Figure 2.21 Log shear stress vs log shear rate (Al-Mulla et al., 2013).	44
Figure 2.22 Log shear viscosity vs log shear rate (Al-Mulla et al., 2013).	44
Figure 3.1 Flowchart methodology.	46
Figure 3.2 Sugar palm fiber.	47
Figure 3.3 Recycled polypropylene.	48
Figure 3.4 NaOH in pallet form.	49
Figure 3.5 Preparation of sugar palm fiber.	51
Figure 3.6 Recycled PP in pellets form.	52
Figure 3.7 NaOH solution.	53
Figure 3.8 Fiber treatment process.	54
Figure 3.9 Electrical siever machine.	55
Figure 3.10 TGA machine model Q50 V20.13 Build 39.	56
Figure 3.11 PerkinElmer (USA) Diamond thermogravimetric (TG)/DSC machine.	57
Figure 3.12 Twin Extruder machine.	59
Figure 3.13 Final product of 3D printing filament material.	59
Figure 3.14 Water Absorption Methodology.	61
Figure 3.15 Tensile Test machine model Instron Universal Testing Machine.	62

Figure 3.16 SEM machine model Zeiss Evo 18 Research.	63
Figure 4.1 TGA curve for recycled polypropylene.	66
Figure 4.2 DSC curve for recycled polypropylene.	66
Figure 4.3 TGA curve for treated sugar palm fiber.	67
Figure 4.4 TGA curve for untreated sugar palm fiber.	68
Figure 4.5 Percentage of water absorption for 24 hours.	70
Figure 4.6 Tensile strength of rPP/treated SPF composites.	72
Figure 4.7 Tensile strength of rPP/untreated SPF composites.	72
Figure 4.8 SEM micrographs of (a) 1% rPP/treated SPF (b) 3% rPP/treated SPF (c)	

5% rPP/treated SPF (d) 1% rPP/untreated SPF (e) 3% rPP/untreated SPF

(f) 1% rPP/untreated SPF composites.

5

74

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF SYMBOLS AND ABBREVIATIONS

PP	-	Polypropylene
SPF	-	Sugar Palm Fiber
PET	-	Polyethylene Terephthalate
3D	-	Three-dimensional
(1)	-	Formula
TPU	-	Thermoplastic Polyurethane
TGA	-	Thermogravimetric Analysis
SEM	-	Scanning Electron Microscopy
HIPS	- 14	High Impact Polystyrene
ABS	and and a second se	Acrylinitrile Butadiene Styrene
AM	-EX	Additive Manufacturing
FDM	1	Fused Deposition Modeling
PLA	ela.	Polylactic Acid
PVA		Polyvinyl Alcohol
PC	ملاك	Polycarbonate in in
MgO	-	Magnesium Oxide
Al_2O_3	UNIVE	Aluminium Oxide KAL MALAYSIA MELAKA
H_3PO_4	-	Phosphoric Acid
PS	-	Polystyrene
HDPE	-	High Density Polyethylene
LDPE	-	Low Density Polyethylene
PVC	-	Polyvinyl Chloride
NaOH	-	Sodium Hydroxide
N_2	-	Natrium gas



LIST OF APPENDICES

APPENDIX	TITLE	PAGE
APPENDIX A	Gantt Chart of Study for PSM 1	84
APPENDIX B	Gantt Chart of Study For PSM 2	85
APPENDIX C	Turnitin Result	86
APPENDIX D	BDP THESIS STATUS VERIFICATION FORM	96
APPENDIX E	BDP THESIS CLASSIFICATION LETTER	97



CHAPTER 1

INTRODUCTION

1.1 Background Study

Presently, polypropylene (PP) is plastic-based material that being widely used for commercial and household purposes along with polyethylene terephthalate (PET) and other thermoplastic material. PP is one of the most affordable plastics available today, and it is used both as a plastic and a fiber in sectors such as automotive manufacturing, furniture assembly, and the aerospace sector. However, PP has become a major threat to the environment due to its non-degradable property. The development of biodegradable polymer from recycling resources has been promoted by increasing environmental awareness to restore conventional non-biodegradable polymer in various applications (Mendes et al., 2016).

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

PP offers several benefits such as low cost, high melting point, sustainable and most important thing is, it is 100% recyclable. Some research have been done using recycled material such as recycled polypropylene reinforced with natural fiber composite to overcome this issue. There have certain interest of using natural fibers as reinforcement in 3D printing filament because there is a growing need for sustainable, save cost, and environment benign resources in a spectrum of uses. Natural fibers were developed with the aim of making lighter composites at cheaper prices than composite material reinforced polymer materials that already exist (Akil et al., 2011). Therefore, petroleum-based materials, such as PP and PET, are used comprehensively with natural fibers, such as sugar palm fiber, palm oil fiber, jute, hemp, and wood dust.

The addition of composites of organic materials and thermoplastics as reinforcement has created more "green" composites and can replaced conventional glass fiber and other synthetic fiber composites that possess manufacturing hazard. This is the effective way to solve environmental issue for waste problem, and production of non-degradable material with the development of environmental friendly material and matrix such as recycled polypropylene reinforced with sugar palm fiber.

1.2 Problem Statement

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The petroleum-based polymer has created a major environmental issues, especially during the disposal stage because of the widespread use by the global society. Global waste production is currently around 1.3 billion tonnes per year, with estimates of 2.2 billion tonnes by 2021 (Khosravani & Reinicke, 2020). In addition, many countries like China have banned plastic bags that can caused many ecological and environmental issue. For example, the most issue caused by plastic bag is the amount of plastic waste produced. Moreover, the most common material used in development of 3D printing filament such as ABS (Acrylonitrile Butadiene Styrene) are one of the factor that can cause environmental issue where it is a hazardous, non-biodegradable substance that, when heated, emits pollutant substances with an unpleasant odour. This shows that 3D printing produced lots of plastic waste to the environment. Hence, to overcome this problem, non-biodegradable polymer need to be replaced with renewable natural sources.

PP is one of the most commonly used thermoplastic material along with PET that can be found in 3D printing filament. It is a biodegradable plastic material that is widely employed in a variety of goods. According to LeBlanc, (2016), because of its melting temperature and hardness, PP is the highest widely used plastic bottles product in the United States, with nearly £5 billion manufactured in 2010. Unfortunately, this will cause the vast majority of these thermoplastic end up as plastic waste in landfills, because of its short lifespan. Moreover, plastic waste in disposal area, PP-based products decompose gradually, taking 20-30 years to totally dissolve. A proper change should be made by recycling this plastic so that it can reduce more waste in landfills and also the properties of PP can be improved. Therefore, it is the most environment conscious and save cost strategy to deal with this issue.

Sugar palm fiber (SPF) was extracted from sugar palm tree (*Arenga Pinnata*) which were usually found in Southeast Asia like Malaysia and Indonesia. There are several parts from sugar palm tree like the fibers, the trunk, the root, its leaves, sap from flowers, and its fruits which can utilize for making many useful products. For example, the fiber from sugar palm tree are suitable for usage as reinforcement material in the making of 3D printing filament. However, SPF has not yet frequently used as reinforcing in the manufacturing of composite materials (Sanyang et al., 2016). This is because the local people did not have enough exposure on 3D printing. They usually utilized the fiber to make brooms, brushes, ropes, door mat, and etc. As a natural fiber, SPF is the key component that become one of the element for reinforment polymer composites.

The project justification of this study is to develop 3D printing filament material using recycled PP reinforced SPF which comes from natural resources to solve the problem of an environmental issue and develop biodegradable polymer. Secondly, to characterize the physical and environmental properties whether the recycled polypropylene and sugar palm fiber substance that is completely recyclable and may be properly rid off in the surroundings.

1.3 Research Objective

The aim of this research is to develop the 3D filament material from recycled polypropylene reinforced with sugar palm fiber. Hence, the objectives are as follows:

- a) To characterize the physical and thermal properties of sugar palm fiber and recycled polypropylene.
- b) To evaluate the effect of sodium hydroxide treatment on the sugar palm fiber thermal properties.
- c) To analyze the physical, mechanical and morphological properties of recycled polypropylene composite reinforced with sugar palm fiber.

1.4 Scope of Research

WALAYS !!

In this study, recycled polypropylene was used as the primary material. Recycled polypropylene was developed from recycled factory in the form of pellets. The physical properties of recycled PP will be tested by using TGA and DSC. Then, sugar palm fiber was added as reinforcement into recycled polypropylene to produced 3D printing filament material. The modification of recycled polypropylene reinforced with sugar palm fiber was performed by inserting different amount of sugar palm fiber loading (0%, 1%, 3%, 5%) into the polymer matrix to find the characteristic of their physical, mechanical and morphological properties. The fabrication process will be using twin screw extruder to form 3D printing filament. Therefore, the application of the reinforcement of sugar palm fiber with recycled polypropylene to develop 3D filament material.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In the industry field, the words "plastic" and "polymer" are often used interchangeably. Plastics are actually chains of molecules linked together which these chains can form into polymers. Plastics may be shaped, stretched, cast, and blasted into virtually any form, sheet, or foam, and they can even be made into textile threads. Over the years, additive manufacturing (AM) which also known as 3D printing is becoming more popular, and new developments are being launched all the time. 3D printing is the technique of layering materials to create a three-dimensional object. In the AM process, there are lots of materials that have been used and they are pure polymers, polymer matrix composites, polymer ceramic composites, nanocomposites, and fiber-reinforced composites. These materials have their own specialties and important factors such as material type, texture, cost, etc. However, plastic waste product from petroleum-based is quite pricey and difficult to recycled. This issue should be solved by producing a biodegradable product made from renewable materials such as recycled polypropylene.

2.2 3D printing in general

After 30 years of 3D printing was invented, additive manufacturing (AM) has progressively outgrown its specialty uses and is helping develop a wide range of manufacturing methods. AM is utilized in a variety of manufacturing industries, including automotive, biomedical, and aerospace. AM is categorized as a multistep process, or a single step process derived by ISO/ASTM 52900. Binder jetting, directed energy deposition, material extrusion, material jetting, and powder bed fusion are the methods classified by additive manufacturing (Sekar et al., 2019).

2.2.1 Commonly used 3D printing technology

There are three types of 3D printing technologies exist based on the materials utilized in the additive manufacturing process such as liquid-, solid-, and powder-based additive manufacturing. Referring to Table 2.1 shows the types of 3D printing based on methods used in additive manufacturing.

Method	Types
Liquid-based additive manufacturing	Stereo lithography (SL), fused deposition
	modeling (FDM), and polyjet.
Solid-based additive manufacturing	Laminated object manufacturing (LOM)
Powder-based additive manufacturing	Powder bed and inkjet head 3D printing (3DP),
*AINO	prometal, laminated engineered net shaping
كل مليسيا ملاك	(LENS), selective laser sintering (SLS), and electron beam melting.
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Table 2.1 Types of 3D printing.

Out of all technology, fused deposition modeling (FDM) is the most often used technology for printing fiber-reinforced polymer composites. Wang et al., (2017) reported that FDM printers run by the controlled extrusion of thermoplastic filaments, as shown in Figure 2.1 Filaments in FDM machine melt into a semi-liquid form at the extrusion nozzle and are extruded layer by layer onto the print bed, where layers join together and form into end product. Printing parameters, including layer thickness, printing orientation, raster width, raster angle, and air gap may all be modified to enhance the performance of printed components. Referring to Table 2.2 shows the advantages and disadvantages of using FDM machine for 3D printer.



Figure 2.1 FDM setup (Wang et al., 2017).

Table 2.2 Advantage and	l disadvantage of FDM	l technology (Wang e	et al.,	2017)
0	<u> </u>			

Advantage	Disadvantage			
The ability to insert many materials at the	The composite materials must be in filament			
same time	form in order to be extruded			
Low-priced, high speed, and easy to use The material that can be used is restricted to				
auwn	thermoplastic polymers with an appropriate			
اوبيوس سيخ melt viscosity ڪل مليسيا ملاك				
Long-lasting and sturdy	It may be difficult to completely remove the			
UNIVERSITI TEKNIK/	support structure applied during printing			

2.2.2 3D printing filament material

Nowadays, there is a lot of 3D printing filament material with different properties and need different temperature to print. Filament of 3D printing is made from thermoplastics feedstock, which plastics or polymer that melt, can be shaped and mold, and solidify when cooled. Plastic is the suitable material for 3D printing material because of its environmentally friendly. FDM printers are commonly used to create plastic products, in which thermoplastic filaments are melt and mold into shape layer by layer. Some of the examples of most used 3D printing filament material are polylactic acid (PLA), acrylonitrile butadiene styrene (ABS), polyvinyl alcohol plastic (PVA), polycarbonate (PC), pure composite such as polypropylene, polyethylene terephthalate, and other several materials. Other than that, there are filaments made of composites mixed with natural fibers in 3d printing. Adding natural fiber to thermoplastic is a reinforcement material to improve the thermal stability of mechanical properties which extend the operating temperature and enhancing material behavior consistency at both high and low temperatures. However, 3D printing innovations have created an opportunity to recycle thermoplastics as 3D printing feedstock material.

Previous study has investigated the feasibility of recycled polypropylene reinforced with natural fiber and gypsum powder for the uses of FDM. A research conducted by Stoof & Pickering, (2017), mentioned that a set of 3mm composite 3D printing filaments with different weight ratios of hemp, harakeke, and recycled gypsum were successfully extruded. The highest fiber content that could be used while still allowing for precise filament production was 30 wt%. Exceeding this percentage caused a sharkskin surface effect, higher strain on the extruder towards its limits, and difficulties with the existing spooling machine. There was no such maximum content found for gypsum composites, and composites containing up to 50 wt% gypsum were easily fabricated. As they get the result of 3mm composite filament, the strongest composite filament obtained from the study was composed of 30% harakeke fiber in a recycled polypropylene matrix. The tensile strength of this composite filament was 41 MPa, and the young's modulus was 3824 MPa. The result that they obtained shows that the mechanical properties of recycled polypropylene as a matrix material was better compared to pure polypropylene. Besides that, composites with 10% to 50% recycled gypsum had a linear decrease in tensile strength while having a slight difference in young's modulus. Based on the result from the study where it leads to a conclusion that recycled polypropylene matrix have the ability to be used as a strong, stiff, affordable, and recyclable 3D printing filament.

2.3 Natural fiber

The potential of each kind of natural fibre to foster advantages such as renewability, sustainability, non-toxicity, biodegradability, outstanding mechanical properties, and low-cost commodity has attracted a lot of interest to the manufacturing industry. Natural fiber may be utilised in three industries: textile, paper, and fabrics, as well as a reinforcing material for composites. Natural fiber can also replace fiber glass in some application such as in composite part in the automotive industry, construction and plastic industry. Figure 2.2 below shows the classification of natural fibers that consists of vegetable (cellulose), animal (protein), and mineral fibers.



Figure 2.2 Categorization of natural fibers (Hasan et al., 2020).

2.3.1 Types of natural fiber

The sugar palm tree is a Palmae family member and a forest species. Sugar palm fiber (SPF) was extracted from sugar palm tree (*Arenga Pinnata*) which were usually found in Southeast Asia like Malaysia and Indonesia. Other name for this tree that familiar to local malaysian is *enau* or *kabung*. Like its name, this tree is known as multifunctional tree species in the world. Sugar, fresh juice, fermented drinks, and syrup were all first produced by the sugar palm tree. Sugar palm tree may be spotted in abundance along rivers in the Malaysia's rural areas in Perak, Negeri Sembilan, Pahang and Melaka. Figure 2.3 shows the sugar palm tree.



Figure 2.3 Sugar palm tree (Arenga pinnata) (Sahari et al., 2013).

While SPF which locally known as *ijuk*, is one of the most common fibers among researchers. Siregar, (2015) reported that *ijuk* can withstand temperatures of up to 150°C and has a flash point of around 200°C. SPF has been reported to have a length of up to 1.19m and a density of 1.26 kg m-3 (Ishak et al., 2013). The range diameter of the fibers is from 94 µm to 370 µm. SPF which is in black/brown color, is sturdy and long-lasting. The treatment of raw material technique separated the stalk from the SPF. After the cleaning process of SPF, they were dried at room temperature. Then, these SPF were cut into two different sizes between 0.1 cm until 0.5 cm for short cut and between 4.0 cm until 5.0 cm for long cut. Ilyas et al., (2018) have reported that SPF offers some benefits over traditional reinforcement fibre recyclability, non-toxicity, abrasiveness, materials such density, as cost. and biodegradability. Figure 2.4 below shows the physical look of SPF.



Figure 2.4 Sugar palm fiber (S et al., 2020).

Jute is a corchorus capsularis bast fiber extracted from corchorus tree. Jute is the most affordable natural fiber and is produced in large quantities. Jute is by far the most common natural fiber used to improve composite materials. On top of that, jute fiber is primarily used in fabrics for packaging a variety of agricultural and industrial products that require bags, boxes, packs, and wrappings. Jute is broadly used because of its low cost

wherever bulky, strong fabrics and stretch-resistant ropes are necessary. Its physical looks are off-white to brown in color and range in length from 1 m to 4 m. They are extracted from the plant's bast or skin. Jute planting generally requires the environment is hot and wet, with temperatures ranging from 20 °C to 40 °C and relative temps of 70% to 80%. Plus, jute requires 5 cm - 8 cm of rainfall during the sowing season. Figure 2.5 shows the image of jute plant, raw jute fiber, jute sticks and woven jute fiber.



Figure 2.5 (a) jute plant, (b) raw jute fiber, (c) jute sticks and (d) woven jute fiber.

Jute has several clear advantages as a natural fiber, including shine and glitter, high tensile strength, low stretchability, medium heat and fire resistance, and long staple lengths. Some of the drawback in jute fiber include thermal stability, crease resistance, stiffness, fiber shedding, and fading when exposed to sunlight.

2.3.2 Application of natural fiber in composite

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Since 100 years ago, natural composites were used by people in the construction of houses in many countries all over the globe. From mixing the husks or sawdust with clay to create coatings, they also combine straws with clay to create a simple composite material. And since then, natural fiber has put a lot of applications such as in indoor furniture, picnic areas, walls, window frames and many more, because of its environmentally friendly and less maintenance. Apart from that, there are several applications of natural fiber with composite on transportation, medical applications, furniture, and etc. Figure 2.6 illustrates the type of applications for natural fiber and synthetic based composites.

Application	Fiber type	Example	
Application in car	Kenaf, jute, coconut, wood fiber	Seats, engine shield, electronic	
interior 📙		device, upholstery, side and	
E		back door panel, seat back, and	
AINT		bumper.	
Application in	Hemp, oil palm, rice husk, jute,	Window frame, panels, textile	
construction,	sisal bagasse, stalk	and yarn, goods, paper, and	
furniture, building and others.	SITI TEKNIKAL MALA	packaging, constructing drains and pipelines.	
Marine and	Glass fiber reinforced plastic	Doors, staircases, chemical and	
mechanical		water tanks, folded plates of	
application		various forms, walls, and panels.	
Aerospace industry	Carbon fiber reinforced plastic,	Engine cowlings, wing skin,	
	Basalt fiber reinforced polymer	main torsion box, antenna	
		dishes, stiffening spars, cockpit,	
		and rotor blades	

Figure 2.6 Applications of polymeric based composites (Pupure et al., 2015).

2.4 Natural fiber Reinforced Composite

Composite is very popular since thousands of years ago. When two or more different materials are mixed together, it can form into composite material. Both materials can work together to develop the composite's different physical and chemical properties. Also, they can improve strength and stiffness of the material. Composite which offers several advantages such as durability, high impact strength, design flexibility, low weight, and low costs, are widely used in the design and manufacture of final products like aerospace structures and electrical equipment.

2.4.1 Sugar palm fiber reinforced high impact polystyrene (HIPS)

Sapuan & Bachtiar, (2012) have studied from previous study about the tensile strength and modulus of SPF reinforced high impact polystyrene (HIPS) with five different fiber loadings (10, 20, 30, 40, and 50%). The tensile modulus increased at the expense of the tensile strength as fiber loading increased in the HIPS matrix, which was characterized by poor fiber-matrix reliability. SEM analysis revealed fiber pullout, indicating poor adhesion between the hydrophilic fiber and hydrophobic HIPS. Bachtiar et al., (2012) proposed some study on mechanical and morphological properties of untreated SPF reinforced HIPS. Referring to (Figure 2.7) and (Figure 2.8) are the results of tensile modulus and tensile strength that were obtained from the study, also Figure 2.9 shows the result of SEM to determine the surface topology of fibers and their distribution in the matrix.



Figure 2.7 Tensile strength of SPF-HIPS composites for different fibre loadings (Sapuan & Bachtiar, 2012).



Figure 2.8 Tensile modulus of SPF-HIPS composites for different fibre loadings (Sapuan & Bachtiar, 2012).





(a)





(c)



Figure 2.9 The fractured surfaces of SPF/HIPS composites after tensile testing for: (a) 10%; (b) 20%; (c) 30%; (d) 40% and (e) 50% (Sapuan & Bachtiar, 2012).

2.4.2 Jute fiber reinforced polypropylene composite

Chestee et al., (2017) have studied about short jute fiber reinforced polypropylene composite on nonhalogenated fire retardants where short jute fibers and PP were mixed with 10, 20, and 30% magnesium oxide (MgO) and aluminium oxide (Al_2O_3) , respectively, and the fiber were soaked in aqueous phosphoric acid (H_3PO_4) . Fire hazards retardancy properties were evaluated, including ignition time, burn size, and overall launching time. Mechanical properties of the composite, such as tensile strength as well as corrosion properties, such as corrosion test, were evaluated. Figure 2.10, Figure 2.11, Figure 2.12, Figure 2.13 and Figure 2.14 show the mechanical properties of magnesium oxide (MgO), aluminium oxide (Al_2O_3) and phosphoric acid (H_3PO_4) fire retardants controlled short jute fiber composites, including tensile strength, tensile modulus, and ductility, as well as flexural strength and bending modulus.



Figure 2.10 Tensile strength (MPa) (Chestee et al., 2017).



Figure 2.12 Tensile modulus (GPa) (Chestee et al., 2017).


Figure 2.13 Bending strength (MPa) (Chestee et al., 2017).



2.5 Polymer as matrix in composite

The term "polymer' is derives from the Greek word which is "poly" means many and "mer" means part. While, polymer in IUPAC is defined by a complex molecule with a high relative molecular mass whose structure primarily consists of the repetitive repeating of units obtained, literally or theoretically, from compounds with a small relative molecular mass. To make a polymer, large number of monomers are chemically bound together by hydrogen bonding (Shrivastava, 2018). Figure 2.15 illustrates bonding of monomer to form a polymer.



Figure 2.15 Monomers are joined together to create a polymer (Shrivastava, 2018).

Generally, polymers have been used in the industry due to their ease of fabrication and usability. There are two types of polymer which are natural polymer and synthetic polymer. Natural polymer are ones easily obtained from plants and animals, while synthetic polymer are ones artificially made by complex chemical processes in a factory or lab. Polymers that are classified as plastics can be divided into 2 groups, thermoplastics and thermosets. Thermoplastic polymers have a linear or branching molecular structure and it can be recycled. For instance, ABS, which most found as polymer in 3D printing, polyethylene, polypropylene, polyvinyl chloride, nylon, acrylic, and teflon. Meanwhile, thermoset polymer has achieved permanent hardness owing to molecular crosslinking and does not soften or flow when heated.

2.5.1 Type of polymer as matrix in composite

The type of reinforcements used in composites is usually used to classify them. These reinforcements are coated in a matrix that keeps everything together. The reinforcements are used to make the composites stronger. Besides that, matrix used are normally in polymer type. The matrix material, polymer matrix composite are made up of several types which are thermoset, thermoplastic, elastomers, nanoparticles, glass, carbon, steel or Kevlar fibers. Thermoset which consist of unsaturated polyester and epoxy while thermoplastic consist of

Polypropylene, Polyethylene Terephthalate, Polycarbonate, Polystyrene, etc. Moreover, polymer have two types which are natural polymer and synthetic polymer.

Natural polymers are materials that exist naturally or are derived from animals or plants. Natural polymers are important to daily life as they are foundation of human structure. Natural polymers include proteins and nucleic acids found in the human body, cellulose, natural rubber, silk, and wool (Shrivastava, 2018). Starch is a natural polymer consist of hundreds of glucose molecules, whereas natural rubber is a polymer derived from a rubber tree's latex. Some latex is synthetic and made from petroleum-based compounds which it used to produce synthetic rubber polymers. Honey would be another example of a naturally occuring polymer that is widely used in daily life. Figure 2.16 displays natural polymers from plants (rubber tree's latex) and animals (honey from bees).



Figure 2.16 (A) Latex from the rubber tree and (B) Honey is an natural polymer from bees (Shrivastava, 2018).

Synthetic fibers are materials that have been created in a lab. These are also referred to as synthetic materials. There are some of the example of synthetic polymers such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamides (nylon), polyvinyl chloride (PVC), synthetic rubber, teflon, epoxy, and a variety of other materials. Synthetic polymers are generally made from petroleum oil in a controlled setting and have a backbone of carbon-carbon bonds. Heat and pressure combined with a catalyst change the chemical bonds that link monomers in, leading them to connect with each other. Synthetic polymers are used in everyday applications that come within the groups of thermoplastics, thermosets, elastomers, and synthetic fibers. Referring to Table 2.3 illustrates some of the abilities of natural and synthetic polymers.

Natural Polymers	Synthetic Polymers			
Occurs naturally	Artificially produced			
Have been in used since millions of years	Have been made significant since the last			
	125 years			
Similar but non-identical repeating	Identical repeating unit			
Natural reaction controls the properties	Highly engineered properties could be			
نيكا مليسيا ملاك	determined by controlling the reaction			
Usually, biodegradable	Some synthetic polymers are			
UNIVERSITI TEKNIKAL	biodegradable MELAKA			
Similar chain lengths of molecules	Chain lengths could be significantly varied			
	based on the reaction conditions			
Backbone could be of carbon, oxygen, and	Backbone is mostly carbon			
nitrogen				
Environmentally friendly	Environmental friendliness			
Limited recyclability	Some of the synthetic polymers could be			
	recycled multiple times			

Table 2.3 Comparison of Natural Polymers and Synthetic Polymers (Shrivastava, 2018).

2.5.2 Polypropylene

Polypropylene (PP), which also known as polypropene, is a thermoplastics polymer that is employed in a variety of purposes. PP is a polymer plastic that belongs to the 'polyolefin' (alkene-derived polymers) family. It is a multifunctional and hard material with many advantageous physical properties, and it is also able to be recycled. This code is represented by a number between 1 and 7 engraved on the bottom of a package and surrounded by arrows forming a triangle, which is beneficial during recycling because it indicates the type of plastic. This ensures that different types of plastic are separated and recycled efficiently. Referring to Figure 2.17, PP's resin identification code is 5. Based on the type of resin used, all plastics, including PET, HDPE, PVC, LDPE, PP, PS and other plastic, have a 'Resin Identification Code/Plastic Recycling Code.



Figure 2.17 The resin identification code for polypropylene (Thomas, 2012).

2.5.3 Recycling of polypropylene

Recently, researches have focused on the possibility of employing recycled plastic fibers as a matrix for the 3D printing filament. Globally, with over 335 million tonnes of plastic are manufactured each year, with just 9% recycled, resulting in increasing plastic pollution (Plastics Europe & Conversio Market & Strategy GmbH, 2019). The ability to recycle a portion of this plastic waste into a matrix that can be utilised for the 3D printing filament opens up an entirely new channel for recycling plastic waste and decreasing global plastic pollution. Since filaments are used for 3D printing polymeric structures, this method of recycling can save several million dollars each year.

Waste made of plastic is classified based on its resin identification code. A recycling code for 3D printed items should be provided, given the variety uses of AM processes and the rising need for 3D printing technology. Referring to Table 2.4 contains information on the ASTM resin identification code. In this case, recycled material is classified by resin code and then crushed into flakes. The flakes can be used as raw material in filament extruders.

Table 2.4 Resin identification code based on ASTM D7611 (Khosravani & Reinicke, 2020).

Recycling No.	Material	Applications			
1	Polyethylene terephthalate	Thermoformed sheet polyester fibers			
	(PET)				
2	High-density polyethylene	Agriculture pipes playground equipment			
	(HDPE)				
3	Polyvinyl chloride (PVC)	Pipis, flooring children's toy			
4	Low-density polyethylene	Plastic bags, containers laboratory			
	(LDPE)	equipment			
5	Polypropylene (PP)	Industrial fibers, auto parts food containers			
6	Polystyrene (PS)	Plastic utensils packaging peanuts			
7	Other plastics (OTHER)	Bottles, headlight lenses safety glasses			

As for the PP recycling process, it involves three (3) stages where the first stage is separate PP from other plastics. This stage require to do separation of PP from being mixed with other plastics and chemicals by using sink-float separation. Second stage is melting. The separated PP will melt to produce a product that the recycler can offer to manufacturing companies. PP have a good melting point about 205°C. The PP is heated in an extruder until it becomes liquid. For the third stage is cooling and forming into pellets. In this stage, recyclers need to cool down the melted PP and then turn it into small pellets so that they can sell it later to manufactures for their manufacturing processes.

2.6 Polypropylene (PP) as matrix in composite

PP as matrix in composite have an outstanding particle size, mechanical properties, thermal and chemical stability, and they are cost-effective. PP has much potential and application that is useful to the industry and environment. Because of its unique ability to be manufactured in a variety ways and for a variety of applications, PP quickly embraced several of the existing synthetic polymers, particularly in the packaging, fiber, and injection moulding industries. There are variety of application of PP. PP is widely used in automotive sector, for example in battery cartridges, trays and beverage holders, bumpers, internal details, orchestral projectors, and gate finishes. PP also found in medical applications including syringes, pill containers, petri dishes, etc thanks too its waterproof properties, as well as durable endurance, ability to be shaped, and t also can withstand steam sterilisation techniques. Other typical applications are appliances, film, plastic furniture, household goods and packaging.

2.6.1 Recycled polypropylene as matrix in composite

Recycled polypropylene has found in various industries as raw material. For example in major renovation waste have been involved into polymer for recycling as cutting-edge structural composites. Ramos et al., (2020) have done a study and introduced geopolymer as a renewable material developed to substitute concreates in infrastructure design. This is because low shear strength and hardened failure characterize strong concreate. In order to improve unstrengthened concreate, the impacts of applying recycled polypropylene fibers to the properties of cement has been introduced.

2.7 Properties of sugar palm fiber

Properties of SPF can be determine through TGA testing and SEM analysis. TGA testing is an analysis to study the mass change, thermal degradation, and thermal stability of composite, while SEM analysis is to study their physical properties.

2.7.1 Thermal properties of sugar palm fiber

The weight loss, thermal degradation, and thermal stability of composite materials are all tested using thermogravimetric analysis (TGA). The previous study conducted by Mohammed et al., (2016) were demonstrate the TGA curve for pure TPU and TPU-SPF composites. Table 2.5 shows the remaining weight ratio of TPU and the corresponding fiber sizes. A slight weight loss was observed for all mixtures under 100 °C, which was responsible for the moisture of water absorption. The composites 250 μ m TPU-SPF (428 °C) and 425 μ m TPU-SPF (425 °C) separated at slightly higher temperature than the 160 μ m TPU-SPF (417 °C), proving that the connection between the fiber and the matrix expanded as fiber size increased, shown in Figure 2.18. All SPF composites dissolved at a lower temperature unlike pure TPU (431 °C), proving a lower fiber-matrix interrelationship.



Figure 2.18 TGA result of pure TPU and TPU-SPF composites (A. A. Mohammed et al., 2016).

Table 2.5 Residual mass % of pure TPU and TPU-SPF composites (Ausama Al-sarraf
Mohammed et al., 2016).

2)/~	نيكل مليست		اويوته		
Course 1 or	Residual Mass (%)				
Samples	RSITI _{726.99} °C KAL	MA 7303.00 °C ME	LAK 7 _{599.16} °C		
Pure TPU	99.69	94.18	7.41		
160 μm TPU-SPF	99.65	94.30	6.11		
250 μm TPU-SPF	99.73	94.46	7.11		
425 μm TPU-SPF	99.76	94.76	6.37		

2.7.2 Physical properties of sugar palm fiber

Physical properties of SPF also can be obtain from SEM analysis. From Mohammed et al., (2016) study, SEM images of pure TPU, SPF fiber, and TPU-SPF composites with fiber sizes of 160 μ m, 250 μ m, and 425 μ m are shown in Figure **2.19**. At different spots, the pure TPU had a nice translucent cracked surface. Furthermore, the TPU-SPF composites had a week fiber-matrix adherence, as holes between the fibers and matrix were recognised, and the fibers were easily slipped out. Figure 2.19 show that fiber sizes of 160 μ m and 425 μ m had bad impacts on fiber-matrix bonding, due to low physical behavior in these composites, possibly due to even more defects on the fiber surface (untreated fiber). However, the composite with 250 μ m had the best bonding to the TPU matrix. This could be due to the appropriate particle size of 250 μ m fiber size for bonding to the TPU matrix, which gives the proper opportunity to boost interfacial bonding and create a smooth texture with greater physical properties.

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Figure 2.19 SEM of (a) pure TPU, (b) SPF, (c) 160 μm TPU-SPF, (d) 250 μm TPU-SPF, and (e) 425 μm TPU-SPF (A. A. Mohammed et al., 2016).

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2.8 Properties of recycled polypropylene

Properties of recycled polypropylene can be determine from thermal and rheological properties. In this study, thermal analysis were study in numerous types of plastic waste to evaluate the optimum value for thermal degradation. TGA was used to investigate the thermal conductivity of the composite and polymer, which was extruded at the same time as the composites.

2.8.1 Thermal properties of recycled polypropylene

A research were conducted by Morales et al., (2021) on thermal analysis of rice husk (RH) and recycled polypropylene (rPP). TGA was used to measure the environmental conductivity of RH fiber, neat rPP, rPP/RH (5 wt.%), and rPP/RH (10 wt.%). Figure 2.20 displays the percentage loss of weight and mass generic in relation to heat for each content. The temperature of the fiber, which accounts for 10% of the weight, is connected to a first phase starting from 50 to 100 °C. The decomposition of hemicelluloses and slight degradation of lignin are linked to the second phase, which has a significant decline at 280 °C. α-cellulose and remaining lignin degradation are linked to the third phase at 340 °C. Between 165 and 175 °C, a slight weight change has been observed in the rPP curves, which is known to possess of defects due to the recycled origins of the thermoplastic. Furthermore, the main destruction of PP occurs between 400 to 490 °C. The rPP/RH (5 and 10 wt.%) curves show aggregated failure behavior. The first step, which occurs between 50 to 120 °C, is referred to moisture content, while the next two reflect fiber element loss, and the final step, which occurs at 460 °C, is referred to matrix degradation. Finally, at 600 °C, residual mass percentages for RH and rPP/RH (5 and 10 wt.%) composites were about 34.45 %, 3.52 %, and 3.88 %, respectively.



Figure 2.20 TGA curve of RH fiber, rPP and rPP/RH composite filaments obtained using TGA (Morales et al., 2021).

2.8.2 Rheological properties of recycled polypropylene

Rheological properties is the study of the interaction between the filler and the matrix. Furthermore, explore the impact of fillers on polymers, rheological properties is essential for simulation, configuring, and design equipment. A study has been done where rheological analysis was performed on a rheometer with a parallel plate shape with a 25 mm of diameter plates at 175 °C. The melt viscosity η (Pa.s) as a method of the shear rate (1/s) was measured using rheometric studies. Referring to (Figure 2.21) and (Figure 2.22), depict shear stress and shear viscosity as against shear rate curves for recycled PP and malleated polypropylene (MAPP), as well as compatible and incompatible materials.



Figure 2.21 Log shear stress vs log shear rate (Al-Mulla et al., 2013).



Figure 2.22 Log shear viscosity vs log shear rate (Al-Mulla et al., 2013).

2.9 Summary

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From the several studies conducted on the sugar palm fiber, recycled polypropylene, natural fiber, composites and natural fiber reinforced composite, and 3D printing filament material, it can be concluded that polypropylene is a good polymer matrix in composite because polypropylene can be recycled to produce new sustainable biodegradable polymer. This is because polypropylene have good melting point, good mechanical properties, high chemical resistance, low density, low cost. Plus, the addition of natural fiber to polymer composites as reinforcement has created more "green" composites and can replaced conventional glass fiber and other synthetic fiber composites. Even though there are precious research reported on the modification of recycled polypropylene as matrix in 3D printing filament, but it is clear from the literature that no work has been done in developing 3D printing filament material using recycled polypropylene reinforced with sugar palm fiber.

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CHAPTER 3

METHODOLOGY

3.1 Introduction

This chapter outlines the research methods used to carry out the study in detail. Research methods presented in this chapter were performed to achieve the objectives of the study. Figure 3.1 shows the flow of the research methodology.



Figure 3.1 The process flow for this study.

3.2 Raw Material

3.2.1 Sugar Palm Fiber (SPF)

SPF was obtained from Hafiz Adha Enterprise which they were the supplier for Industri Enau Malaysia in Kuala Jempol, Negeri Sembilan, as shown in Figure 3.2. SPF was extracted from its tree in form of thread or strip.



Figure 3.2 Sugar palm fiber.

3.2.2 Recycled polypropylene (rPP)

rPP was obtained from San Miguel Plastic Film Sdn. Bhd in Melaka. Referring to Figure 3.3, shows the matrix in the form of pellets. The product specification of the matrix was shown in Table 3.1.

Table 3.1 Specification of recycled polypropylene.

Specification of recycled polypropylene			
Color	White		
Form	Pellet		
Туре	Virgin/recycled		



Figure 3.3 Recycled polypropylene.

3.2.3 Sodium Hydroxide (NaOH)

In this study, NaOH solution in form of pallet, as shown in Figure 3.4 was used for the alkaline treatment. The material specification for NaOH was shown in Table 3.2.

Items	Specifications
Physical Form	Pellets
Color	White
NaOH Content	More than 99%
Water Solubility	100%
Molecular Weight	40 g/mol

Table 3.2 Specification of Sodium Hydroxide.



Figure 3.4 NaOH in pallet form.

3.3 Preparation of material

3.3.1 Preparation of sugar palm fiber

In order to get a 125 μ m of SPF, there are several steps that have been taken. First and foremost, the SPF need to be clean to remove the contaminant. Using tap water, the SPF were cleaned and washed to separate any impurities such as dust and dried leaves. Then, the

SPF were dried naturally at room temperature for 24 hours after the cleaning process was completed. The next step is cutting process where the dried SPF were cut into small size about 1 mm to 5 mm. The SPF need to be cut into smaller size to avoid from being stucked in the crusher machine and for a smooth process of crushing so that 125 µm of SPF will be achieved. After finish with the cutting process, the SPF were grounded by inserting it gradually into the crusher machine. To avoid overheated, the maximum operation can only take 5 minutes per operation. Cool down the machine at least 10-15 minutes before repeated uses. This process took 1 hour to produce roughly 90 g of 125 µm of SPF powder and only required 75 g of SPF powder for TGA and rheological testing. The particle SPF was sorted in order to acquire 125 µm of particle size. In this process, electrical siever machine were used to sieve the SPF with the mesh size of 125 µm. The excess of the fiber were inserted again into the crusher machine to crush and repeat the sieve process until the required amount of SPF powder is fulfilled. The fiber then packed in a ziplock bag and ready for fiber treatment. Figure 3.5 shows the step by step in preparation of sugar palm fiber.

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3.3.2 Preparation of recycled polypropylene (rPP)

Referring to Figure 3.6, rPP is obtained from San Miguel Plastic Film Sdn. Bhd in Melaka and it was already come in pallet form and is ready to use for TGA testing.



Figure 3.6 Recycled PP in pellets form.

3.4 Fiber Treatment

There are two processes involved in fiber treatment which are the preparation of NaOH solution and the preparation of SPF treatment. Figure 3.8 below will show the overal process in fiber treatment.

3.4.1 Preparation of NaOH solution

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Alkaline treatment using NaOH is a chemical analysis which is it commonly used to improve the fiber properties such as thermal properties to improve the addition between fiber-matrix and it used to remove impurities on the fiber interface (Radzi et al., 2019a). In this study, preparation of NaOH solution consist of 6% the concentration of NaOH and 94% of H_2O will be add together to fulfill the 100% of mixture for NaOH solution. A formula was used to calculate the amount of 6% NaOH using formula in (1).

$$\frac{6}{100} \times 1000 \ g = 60 \ g \ of \ NaOH \tag{1}$$

Meanwhile, the weight ratio for wateris shown in Table 3.3. Both NaOH and H_2O will be stirred together until the solution is dissolved. Referring to Figure 3.7, NaOH solution were dissolved with 6% of NaOH and 96% of H_2O mixture.

No	Material	Amount (g)	Amount (%)		
1	NaOH	60	6%		
2	Water	60	94%		
3	Total	120	100.00%		

Tab	le 3.3	8 W	'eight	t ratio	of	water
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Figure 3.7 NaOH solution.

3.4.2 Preparation of fiber treatment (SPF)

In this process, SPF will be immersed completely in NaOH solution and soaked for 2 hours at room temperature. After 2 hours, the SPF will be rinsed with tap water in a strainer for at least 2 times. Then, the fiber will be crushed into a aluminium foil and dried it in the oven at 104 °C for 24 hours. The treated SPF will be used for testing purpose.



Figure 3.8 Fiber treatment process.

3.5 Characterization of sugar palm fiber (SPF)

In this study, we need to ensure that the characterization of SPF is good enough as reinforcement material to rPP. Particle size analysis and TGA test were conducted to identify and characterize SPF properties in terms of physical and thermal properties.

3.5.1 Particle size analysis

The purpose of this analysis is to measure particle size of SPF. The methods used in this study is siever. The particle size required in this study is $125 \,\mu$ m.

3.5.1.1 Siever

Siever is a traditional methods used to measure particle size of material in powder, fluids, solvents or other types. Crushed SPF was inserted in the electrical siever machine to sieve until it get the size of 125 μ m. Since the particle size of SPF required in this study is 125 μ m, so 100 μ m and 125 μ m were used repeatedly until 125 μ m of SPF can be obtained. Figure 3.9 below shows the siever machine used in this study.



Figure 3.9 Electrical siever machine.

3.5.2 Thermogravimetric analysis (TGA)

TGA is the best extensively used technology for thermal analysis to characterize materials by determine their mass changes as a temperature dependent variable. The thermal properties of SPF was tested by using TGA machine (Q50 V20.13 Build 39), as shown in Figure 3.10. Alkaline treatment have been reported to affect the thermal properties of SPF

(Jumaidin et al., 2017). A study have been conducted by Safri et al., (2020) where they stated that the temperature range was set between 25 °C and 550 °C, with a heating rate of 10 °C/min in a nitrogen flow (N_2) with a flow rate of 50 ml/min. Referring to Akhtar et al., (2016), TGA curves of treated and untreated kenaf fibers were captured in a N₂ flow from room temperature to 600 °C at a heating rate of 10 °C/min. Also, the melting point and maximum temperature limit of the fibers were determined using a Mettler Toledo differential scanning calorimeter 822e.



Figure 3.10 TGA machine model Q50 V20.13 Build 39.

3.6 Characterization of recycled polypropylene (rPP)

As previously mentioned, thermoplastic polymers such as PET and PP account to roughly 90% of all plastics worldwide, but the biochemical characteristics of 3D printed produced items constructed of these semi-crystalline thermoplastics are poorly understood. As PP has lately attracted a lot of interest in 3D printing processes, the material can be recycled and reused if the part was not 3D printed perfectly, or if some defects were appeared to the printed part. Hence, AM field have decided to promote recycled thermoplastic material such as rPP. In this study, rPP have been chose as matrix composite for 3D printing filament material reinforced with SPF. A test have been conducted which is analysis TGA and DSC.

3.6.1 Thermogravimetric Analysis (TGA)

TGA was conducted in order to determine the thermal properties of rPP using TGA instrument model Q50 V20.13 Build 39 as Figure 3.10. Vidakis et al., (2021) reported that to identify the thermal properties of PP after the various recycling rounds, TGA test were conducted in an oxygen air at temperature range of room temperature between 30 °C to 800 °C at a heating rate of 20 °C/min in a N₂ flow with flow rate of 50 ml/min. About 8 mg of sample weight were used in this study.

3.6.2 Differential Scanning Calorimetry Analysis (DCS)

DSC analysis was undertaken to obtain information on thermal transitions that occur in the sample using a Mettler DSC analyzer, as Figure 3.11. Under a dynamic nitrogen flow rate of 50 ml/min, 11 mg of sample weight was performed with heating rate 20 $^{\circ}$ C /min, from 30 to 800 $^{\circ}$ C.



Figure 3.11 Mettler DSC machine.

3.7 Preparation of composite

For development of 3D printing filament material, the mixture of rPP and SPF with a proper fiber loadings will prepare using Labtech Twin Extruder 26 mm as in Figure 3.12 to form composite in form of filament material. Figure **3.13** shows the process of fabrication of rPP reinforced with SPF filament.

3.7.1 Mixing of sugar palm fiber and recycled polypropylene

To form pellet/filament material in this study, rPP will mix with SPF. The modification of rPP reinforced with SPF was performed by inserting different amount of SPF loadings (0%, 1%, 3%, 5%) into the polymer matrix. The mixture of rPP and SPF was prepared manually.

3.7.2 Twin Extruder

After the process of mixing, the blend of rPP and SPF was forced into a mold cavity with high pressure through a sprue. The mixture is fed through a hopper, then conveyed forward to cylinder barrel. The material will be heated to melt and soften the composite. The material may be chilled and solidified in a organised way thanks to the cooling system in the mould. The composite is held in the mould until it hardens, at which time the mould opens and the component is removed with the help of ejected pins. Then, the composite material formed filaments for 3D printing.



Figure 3.12 Twin Screw Extruder machine.



Figure 3.13 Fabrication of 3D printing filament material.

3.8 Characterization of composite

To enhance the composite of rPP and SPF, the filaments then will be analyse to identify its physical, mechanical and morphological properties through three different testing, including water absorption, tensile test, and SEM.

3.8.1 Physical Properties

Water absorption test is one of the most crucial physical attributes to consider how much water has been absorbed between the filament. Figure 3.14 shows the water absorption process methodology.

3.8.1.1 Water Absorption

Thirty samples of filament was prepared with a 20 cm long each for water absorption test and were stored in a vacuum bags a day before to absorb any moisture, since this is the only way of ensuring that the filament itself will be kept dry. After dry the sample, the initial weight of the sample, (W_i) before immersed was weighed. Then all the sample was soaked in 20 ml distilled water for 24 hours and kept it in a room. After 24 hours, the samples were took out from the container and the remaining water was removed. The sample will be weighed (W_f) . The water absorption of the sample was calculated by using the following equation.

Water absorption
$$\% = \frac{W_f - W_i}{W_i} \times 100\%$$

Where, W_i represents the initial weight of sample before immersion and W_f represents the final weight of sample after 24 hours of immersion in distilled water.



3.8.2 Mechanical Properties

The mechanical properties for rPP/SPF filament were determined from tensile test. The purpose of tensile test for 3D printing filament is to determine how it will behave under load. Approximately 18 samples of the filament were tested for their tensile properties and an average was recorded.

3.8.2.1 Tensile Test

The filament was cut with an overall length of 200 mm for the tensile test. Tensile test can be done by using Instron Universal Testing Machine as shown in Figure 3.15. According to Lomelí Ramírez et al., (2011), the tensile sample of Cassava starch-green coir

composite were tested at ambient temperature and relative humidity (RH) of 75% at a testing speed of 5 mm/min.



Figure 3.15 Tensile Test machine model Instron Universal Testing Machine.

3.8.3 Morphological Properties

Morphological test can be done using scanning electron microscopy (SEM) to determine the characterization of composite in terms of surface topology. Figure 3.16 below show the SEM machine, model type Zeiss Evo 18 Research, in order to get SEM micrographs taken from 6 fractured tensile test samples of rPP/SPF filament. All of the samples were coated with gold before being observed by sputtering in a vacuum tank before undergo morphological test (Lomelí Ramírez et al., 2011).



Figure 3.16 SEM machine model Zeiss Evo 18 Research.

3.9 Summary

In summary, this chapter discuss the method proposed for developing a new 3D printing filament material using recycled polypropylene reinforced with sugar palm fiber. The main goal for this study is aim to succeed and achieved the objective of the study. Equally important, to achieve particle size of SPF 125 µm, SPF have undergo several processes from preparation of raw SPF to sieve process. Then, SPF need to undergo alkaline treatment using 6% of NaOH solution where the SPF will be immersed completely in NaOH solution for 2 hours. After 2 hours of immersion process in NaOH solution, the SPF will be dried in the oven for 24 hours. Nevertheless, rPP already obtained in pellets form. Furthermore, both rPP and SPF (treated and untreated) were sent to lab for TGA analysis. Aftermost, to develop 3D printing filament, rPP and SPF will be mix together with different fiber content (0%, 1%, 3%, 5%) accordingly and will be forced into twin extruder machine until formed 3D printing filament (Figure 3.13).

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

This chapter presents the results of the recycled polypropylene (rPP) reinforced with sugar palm fiber (SPF) by using thermal, physical, mechanical and morphological testing. Different parameter of SPF was used to determine the effect on the polymer matrix used which is rPP. The outcome of the graph was then analyzed and discussed for thermal, physical, mechanical, and morphological testing of the samples. The result is relevant to the intent of this paper.

4.2 Thermal Properties

Thermal analysis testing was performed to calculate the degradation curve of the raw materials such as rPP, treated SPF and untreated SPF towards their degradation temperature (°C), using thermogravimetric analysis machine (TGA) and Differential Scanning Calorimetry (DSC). Thermal analysis is important to determine the thermal properties of the composite. The melting temperature, thermal breakdown, and weight loss of rPP, treated SPF and untreated SPF were examined between 30 °C to 800 °C at a heating rate of 20 °C/min in nitrogen gas (N₂) flow at 50 mL/min.

DSC analysis will be done to obtain information on heat transfer reactions in the sample. Changes including residual sensitivity, moisture absorption, melting, crystallization, particle change, and glass transition temperature. Differential Scanning Calorimetry (DSC) analysis was conducted with heating rate $20 \degree C / min$, from 30 to 800 $\degree C$. The sample weight

that was used in this study was about 11 mg. (Figure 4.1 to Figure 4.4) below shows the typical curves pattern of TGA and DSC on rPP, TGA on treated SPF and untreated SPF.

4.2.1 TGA and DSC result for recycled Polypropylene

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In Figure 4.1 shows the TGA curve for rPP where the phase of rPP at first quartile degradation occur at 370.80 °C. Also, a small amount of mass was dropped which can be seen at 100.4%. During the second quartile of degradation occur at 430 °C took place with all the material were fully degraded. This is because at this stage, at its most extreme, weight reduction can be noticed, which the molecule bonding for rPP has completely decompose at higher rate.

Furthermore, Figure 4.2 shows the heat flow curve of the DSC result was obtained to evaluate the rPP melting temperature, T_m where it shows an endothermic events at 167.94 °C. It was attributed to melting point of rPP. The maximum melting temperature for rPP is 472.71 °C. Thus, from these results, the temperature setting for twin screw extruder machine can be determined between 160 °C to 190 °C.

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Figure 4.2 DSC curve for recycled polypropylene.
4.2.2 TGA results for treated and untreated SPF

The thermogram in Figure 4.3 shows that at the first quartile degradation of treated SPF, the initial transition start at 29.88 °C until 264.63 °C with weight loss of 8.125% of fiber content (moisture). This is because, the decrease in percentage weight can caused by the loss of moisture content in fiber (Nazrin et al., 2020). The second phase of degradation start at 264.63 °C until 331.70 °C with 17.94% of mass loss. This is due to the decomposition of other material inside the fiber component, such as hemicellulose and lignocellulose (Izwan et al., 2021). Final phase of degradation occur at 331.70 °C resulted in a greater mass loss 52.10% formation of ashes. While in Figure 4.4 for untreated SPF curve patterns, the initial decomposition occur at temperature 98.35 °C - 233.32 °C (2.55% weight loss) which most of the component that degrade moisture content. However, the second phase of degradation start at 310.76 °C until 365.46 °C where lignocellulos and hemicellulose content were decomposed. For the final degradation starting at 457.81 °C and started to form ashes.



Figure 4.3 TGA curve for treated sugar palm fiber.



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A research study conducted by Radzi et al., (2019b) stated that water is an issue for hybrid composites made from natural fiber sources such as SPF (hydrophobic or hydrophilic). Therefore, water absorption test is one of the most essential measurements for determining the amount of water absorption and as a result of this study's methodology.

4.3.1 Water absorption test

4.3

Water absorption is a way to analyze how much water is absorbed under specific conditions. The weight, void/pore, viscocity of matrix used, fiber content ratio and diameter of the filament are all factors that can affect water absorption. A water uptake test was performed to see how much water was taken between the samples of filament exposed to

water based on its fiber loading. Referring to Lomelí Ramírez et al., (2011), water absorption for thermoplastic starch-based materials is essential since the material's characterization can influence its efficiency while also helping to save the planet. Owing to its hydrophobic nature, it was noticed that the rPP/untreated SPF composite received the least amount of water or moisture. Nevertheless, it showed that in the rPP/treated SPF composite, there was a slightly increase in water absorption percentage due to hydrophilic nature.

Figure 4.5 shows the water absorption percentage of rPP/SPF filament. It can be seen that after 24 hours of immersion in distilled water, the 5% of fiber loading for treated SPF shows 4.66% of the highest water uptake, followed by 3.49% (3% treated SPF) and 2.75% (1% treated SPF). Based on the observation, the water uptake for rPP/treated SPF shows a steady increase when more SPF fiber loadings were added with rPP. Moreover, the greater the amount of fiber loadings used, the higher the water absorbed. This is due to lots of natural fiber use, more water absorbent. The thickness of the fiber, the existence of holes, and the adhesion between fiber-matrix itself are indeed correlated to the ability of water uptake (Nunna et al., 2012). These factors resulted to water becomes trapped in the void that formed inside the filament, has caused to increasing in composite weight. Eventhough the SPF had undergo alkaline treatment using 6% of NaOH solution, it shows that NaOH treatment cannot reduce the hydrophilic nature of the fiber which leads to increase the water uptake.

However, for the rPP/untreated SPF filament shows a steady decrease in water uptake with the addition of fiber loadings. The highest water uptake at 1% of fiber loadings shows 12.59% of water absorption, followed by 3% of fiber loadings at 7.71% of water absorption, and 5% fiber loadings shows the reduction of water uptake at 4.49%. According to this trend, it is clear that as the addition of fiber loadings to rPP/untreated SPF increased, the amount of water absorbed decreased. This is resulted to less void appeared inside the filament, so

less water absorbent. Even without the alkaline treatment, lignocellulosic structure such as hemicelluloses in the SPF tends to be hydrophobic, so it results to lower water uptake (Yew et al., 2019).



4.4 Mechanical Properties

The mechanical properties of rPP/SPF hybrid composites are significant in a lot of uses. The behaviour of the composites are relying on the bonding between fiber-matrix. There are factors that will affect mechanical properties such as fiber loadings, element percentage, fiber-matrix adherence, interaction between fiber and matrix, and process parameters (Rashid et al., 2017). 18 samples of the rPP/SPF filament were tested and graph of tensile strength were discussed further.

4.4.1 Tensile Test

Figure 4.6 and Figure 4.7 below shows the graph impact of NaOH treatment on tensile test for rPP/SPF hybrid composites. As a result of the tensile strength, the mechanical properties of rPP/treated SPF hybrid composites have changed. The structure or surface of treated SPF reduced the tensile strength even after the lignin and hemicellulose components were withdrawn. This illustrates that when compared to the rPP/untreated SPF, the result of NaOH treatment on treated SPF leads to a slightly decreased in tensile strength, as shown in Figure 4.6. The tensile strength of rPP/treated SPF hybrid composites dropped with the addition of fiber loading percentage. It is clearly that the NaOH treatment, on the other hand, may cause some damages to the SPF structure or surface. This can have an impact on the composites was recorded with the least tensile strength value at 9.29949 MPa. NaOH treatment has proven to weaken the filament (Halápi et al., 2018). This is due to lot of voids appeared inside the filament, which has caused the filament to brittle and easily broken.

However, Figure 4.7 shows the different trend where the tensile strength of rPP/untretaed SPF hybrid composites shows a steady increased. From the graph, it is observed that 5% of fiber loadings have the highest tensile strength value than 0% of fiber content in pure rPP filament (16.8133 MPa), which at 21.8902 MPa. This is because strong bonding and good adhesion between untreated SPF and rPP makes it the highest among other samples. Other several reasons, including the rPP/untreated SPF have improved wettability, fewer void, and decrease in micro-cracking (Radzi et al., 2019a). Besides, it seems that without NaOH treatment, the tensile strength for the filament has improved with increase in fiber loadings.



Figure 4.6 Tensile strength of rPP/treated SPF composites.



Figure 4.7 Tensile strength of rPP/untreated SPF composites.

4.5 Morphological Properties

The most well-known and commonly used in morphological properties is scanning electron microscopy (SEM). Thus, in this study the SEM analysis are used to justify the surface topology of the cross-section area on filament.

4.5.1 Scanning Electron Microscopy Analysis (SEM)

SEM as described by Mendes et al., (2016), is an evaluation of qualitative properties of the material. The internal surfaces and cross-section for the filament were commonly evaluated through the SEM in order to investigate the good adhesion of the two or more materials used (Sahari et al., 2013). SEM analysis was done to evaluate how adequately the SPF mixed with the rPP and to assess the surface structure.

Figure 4.8 shows the morphologies of rPP/SPF hybrid composites at 500x magnification. The majority of the samples from the rPP/SPF filament spool were nicely mixed, according to the SEM micrographs. Based on the SEM image obtained in Figure 4.8(a) below, it can be observed that the 1% of rPP/treated SPF filament have less void compared to 5% of rPP/SPF filament, which in Figure 4.8(c). This happened when the amount of fiber loading increased, more fiber is well attached and large voids are visible at the cross-section area. Possibly, this might have been the cause of poor in tensile strength. The NaOH treatment, on the other hand, definitely causes some damage to the SPF structure, which might impact the composites' mechanical properties (Radzi et al., 2019a). In fact, the filament surfaces that have been modified with NaOH were rougher than untreated fiber composites.

Furthermore, in Figure 4.8(d), the internal surface of cross-section area in 1% rPP/untreated SPF filament was consistent, fine, and free of damaging surface imperfections.

Also, fiber cannot be observed clearly, resulted to high tensile strength. While, in Figure 4.8(e) and Figure 4.8(f) shows a good adherence between fiber and matrix. This factors are proved to the high tensile strength of the filament. Thus, without treatment with NaOH, the tensile strength of rPP/untreated SPF filaments has improved. This is due to high hydrophilicity, less porosity, and strong fiber-matrix adhesion.



Figure 4.8 SEM micrographs of (a) 1% rPP/treated SPF (b) 3% rPP/treated SPF (c) 5% rPP/treated SPF (d) 1% rPP/untreated SPF (e) 3% rPP/untreated SPF (f) 1% rPP/untreated SPF composites.

4.6 Summary

Basically, as determined in this chapter from water absorption testing results, which owing to hydrophobic effect, it can be summarized that the water absorption percentage for rPP/untreated SPF composites decreased. This is due to less voids or gaps existed in in the filament. Despite this, the hydrophilic effect on the rPP/treated SPF resulted in an increase in water absorption percentage, as illustrates in Figure 4.5. In addition, the mechanical properties can be observed through tensile testing as showns in Figure 4.6 and Figure 4.7. The tensile strength of rPP/treated SPF hybrid composites decreased, caused by NaOH treatment as it proven to weaken the filament. Besides, it seems that without NaOH treatment, the tensile strength for the rPP/untreated SPF filament has improved with increase in fiber loadings. Indeed, from SEM images as represented in Figure 4.8 shows that there are large voids existed in the 5% of rPP/treated SPF composites and less void appeared in the rPP/untreated SPF composites. Last but not least, the results obtained from TGA analysis determined the thermal properties behaviour of rPP, treated SPF and untreated SPF. It is essential to predict the percentage weight of degradation for SPF after undergoing alkaline /FRSITI TEKNIKAL MALAYSIA MELAKA treatment, and for rPP after the various of recycling process.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

Many various studies have been done using polypropylene as thermoplastic as it is a viable element for developing environmentally friendly products and biodegradable material. However, polypropylene has limitations as it often affected by UV degradation, it has limited use in high temperature which can lead to oxidisation, polypropylene also once heat is applied, they have weak adhesion characteristics and are highly combustible. Because of these drawbacks, polypropylene filament is not easily accessible for 3D printing (Staff, 2016). Thus, the modification techniques were applied where the recycled polypropylene were used to upgrade the properties of this material.

Based on the research objectives described here in section 1.2, the following are the overall results from this research paper:

Objective 1: Characterize the physical and thermal properties of sugar palm fiber and recycled polypropylene.

Sugar palm fiber and recycled polypropylene were characterized by physical and thermal properties. This project used siever to obtain 125 µm particle size of SPF, while rPP already come in pellet form. After that, some part of SPF were undergoing alkaline treatment to enhance its fiber properties such as thermal properties and to remove impurities from the SPF fiber surface. Then, both SPF and rPP were sent to lab for thermal testing using TGA analysis.

Objective 2: Evaluate the effect of sodium hydroxide treatment on the sugar palm fiber thermal properties.

To promote natural fiber and polymer matrix surface adhesion as in this case is using recycled polypropylene reinforced sugar palm fiber, alkaline treatment are the most common used among all researcher. Alkaline treatment used in this project is 6% of sodium hydroxide solution (NaOH). Moreover, NaOH treatment is proved to improved the thermal and physical properties of SPF. However, rPP/treated SPF hybrid composites shows worse tensile strength because from the tensile test result, the tensile strength graph shows slightly decreased.

Objective 3: Analyze the physical, mechanical and morphological properties of recycled polypropylene composite reinforced with sugar palm fiber.

The physical properties of rPP reinforced with SPF composite (water absorption) shown a different trend where the graph for water absorption on treated SPF is increased while the graph for water absorption on untreated SPF decreased. Other than that, the mechanical properties of rPP reinforced with SPF were analyzed through a tensile test. It can be observed that rPP/untreated SPF composites have a good tensile strength compared to rPP/treated SPF composites. For the morphological properties of rPP/SPF hybrid composites were analyzed using SEM analysis. SEM micrograph shows a good adhesion between SPF and rPP, but more voids are visible on 1%, 3% and 5% of rPP/treated SPF sample.

5.2 **Recommendations**

For future improvements, the development of 3D printing filament material using recycled polypropylene reinforced with sugar palm fiber could be enhanced as follow:

- i. First and foremost, carry out more research on sugar palm fiber properties as the reinforcement for polymer matrix in developing 3D printing filament material.
- Alternatively, suggestion to try other recycled thermoplastic and other natural fiber such as recycled Polylactic Acid, recycled Polystyrene, recycled Polyethylene, recycled ABS, kenaf fiber, jute fiber, and roselle fiber because these materials are easily available in Malaysia.
- iii. Furthermore, recommend to use other alkaline treatment such as silane because NaOH treatment on sugar palm fiber seems to not improved the mechanical properties of treated sugar palm fiber because the result from tensile test shows decreased in tensile strength.

5.3 Project Potential

In general, this project potential have a several which are:

- i. This project using recycled polypropylene reinforced with sugar palm fiber can be extrude using twin extruder because both material have a suitable temperature for 3D printing filament material.
- ii. Furthermore, this 3D printing filament material are suitable to print for 3D model as a new advancement to replace commonly used 3D printing filament such as ABS.
- iii. Equally important, this project is feasible relate since it is safe and great for environments because recycled polypropylene are non-toxic plastic and it is suitable for environmental surrounding.

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APPENDICES

APPENDIX A Gantt Chart of Study for PSM 1

ACTIVITIES	STATUS	WEEK															
ALA	AYSIA	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
SUPERVISOR SELECTION AND REGISTERED TITLE	PLAN	1.															
25	ACTUAL	8															
BRIEF AND PROJECT EXPLANATION BY SUPERVISOR	PLAN	17															
2	ACTUAL																
MODULE #1 RESEARCH DESIGN AND PLANNING	PLAN							·									
F	ACTUAL																
DISCUSS PROBLEM STATEMENT AND OBJECTIVE FOR	PLAN																
CHAPTER 1	ACTUAL				-												
MODULE #2 FINAL YEAR PROJECT LITERATURE	PLAN				1			1	1								
REVIEW	ACTUAL																
DRAFTING LITERATURE REVIEW AND WRITING UP	PLAN																
CHAPTER 2	ACTUAL		1			1											
MODULE #3 RESEARCH METHODOLOGY	PLAN	0					3.5		1.11		A. 13						
	ACTUAL	0						15		V.	7	7					
RESEARCH ON METHODOLOGY AND WRITING UP	PLAN							1									
CHAPTER 3	ACTUAL																
WRITING UP PRELIMINARY RESULT	PLAN	TEP	(N)	KΔ		AL	_A`	'SI	ΑN	1EI	AP	(A)					
	ACTUAL																
SUBMISSION OF FIRST DRAFT PSM 1	PLAN																
	ACTUAL																
SUBMISSION OF SECOND DRAFT PSM 1	PLAN																
	ACTUAL																
PREPARATION AND PRESENTATION PSM 1	PLAN																
	ACTUAL																

APPENDIX B G

3	Gantt Chart of Study For PSM 2	
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ACTIVITIES	STATUS	WEEK															
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
MEETING AND DISCUSSION	PLAN																
	ACTUAL																
PREPARATION OF RAW MATERIAL	PLAN	100															
	ACTUAL	200															
CONDUCTING THE EXPERIMENT	PLAN	1															
S	ACTUAL	1															
COLLECT DATA AND MAKE ANALYSIS ON SAMPLE	PLAN																
ш.	ACTUAL																
DISCUSS ON RESULT EXPERIMENT	PLAN																
5	ACTUAL								_								
START DRAFT REPORT AND WRITING UP CHAPTER 4	PLAN																
d'a	ACTUAL				1				1								
START DRAFT REPORT AND WRITING UP CHAPTER 5	PLAN																
	ACTUAL																
SUBMISSION OF FIRST DRAFT PSM 2	PLAN		1			1						1					
2010	ACTUAL	a 1	-				2.15		h. d. h	1. 1	6.13	4					
RECHECK FIRST DRAFT	PLAN	0						15		1	1	2					
	ACTUAL							1		-							
WRITING UP CONCLUSION FOR THIS STUDY	PLAN	a															
UNIVER	ACTUAL	TEP	(NI	KA		IA	LAY	rst	ΑŇ	TEL	AP.	A					
SUBMISSION OF FULL REPORT	PLAN																
	ACTUAL																
FINALIZE THE CORRECTION OF FULL REPORT	PLAN																
	ACTUAL																
PREPARATION AND PRESENTATION OF PSM 2	PLAN																
	ACTUAL																