HYGROTHERMAL AGEING OF RECYCLED POLYPROPYLENE AND RUBBERWOOD WASTE COMPOSITE FOR CONSTRUCTION MATERIAL

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UNIVERSITI TEKNIKAL MALAYSIA MELAKA
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Submitted in accordance with the requirement of the Universiti Teknikal Malaysia Melaka (UTeM) for a bachelor's degree in Manufacturing Engineering

(Hons.)

by

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FACULTY OF MANUFACTURING ENGINEERING

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DECLARATION

I, at this moment, declared this report entitled “Hygrothermal Ageing of recycled polypropylene and rubberwood waste composites for construction materials” is the result of my research except as cited in references.

Signature : ……………………………………

Author’s Name : DALGUSH DIPAK AJANG

Date : 23 JUNE 2022

APPROVAL

This report is submitted to the Faculty of Manufacturing Engineering of Technical University of Malaysia Melaka as partial fulfilment of the requirement for a Degree of Manufacturing Engineering (Hons). The member of the supervisory committee is as follow:

………………………………………………

(PROF. MADYA DR. ZALEHA BINTI MUSTAFA)

ABSTRAK

“Kajian ini bertujuan untuk membandingkan sifat fizikal, sifat lenturan dan menilai permukaan rekahan komposit kayu getah(RWF) diperkuat polipropilena kitar semula(rPP) disebabkan penuaan higroterma. RWF bersifat hidrofilik, menyerap air, dan sifat rPP berubah apabila terdedah suhu tinggi. Kelembapan menyebabkan permukaan antara komposit polimer kayu(WPC) merosot, mengurangkan sifat lentur. Bahan dengan komposisi 37.5wt.% RWF(pengukuh), 50.3wt.% rPP(matriks) dan 12.2wt.% aditif ditekan mampat dengan haba dan kemudian dimesin mengikut ASTM D790. Kemudian, penuaan higroterma dilakukan selama 30 hari pada 50°C dan 90% kelembapan relatif(RH) untuk kaedah pertama dan dalam rendaman air pada 50°C untuk kaedah kedua. Tingkah laku penyerapan air mengikuti tingkah laku Fickian. Kadar penyerapan dalam rendaman air(0.96m^2/s) lebih tinggi daripada 90%RH(0.95m^2/s). Peningkatan tebal menunjukkan kestabilan dimensi komposit lemah dalam persekitaran yang lembap. Kekuatan awal komposit ialah 30.49Mpa mengurangkan selepas terdedah kepada wap(27.85Mpa) dan lebih rendah selepas terdedah kepada rendaman air(26.61Mpa), sama dengan corak modulus menunjukkan kelembapan merosakkan kekuatan lentur dan modulus komposit. Tegangan pada patah komposit meningkat selepas terdedah, menunjukkan komposit menjadi mulur disebabkan oleh kesan pemplastikan rPP. Sampel tidak terdedah diperhatikan gagal kerana gentian dan matriks patah, menyebabkan keretakan rapuh. Apabila terdedah kepada wap 90%RH dan rendaman air, gentian tercabut dan penambahan retakan mikro dan jurang antara gentian dan matriks dapat dilihat. Ia menunjukkan bahawa antara muka gentian dan matriks menjadi lemah. Konklusinya, penuaan higroterma mengubah dimensi komposit dan mengurangkan sifat lentur dengan imej SEM untuk menyokong kesan akibat penuaan higroterma.”

ABSTRACT

This study aims to compare the physical and flexural properties and evaluate the fracture surface of rubberwood(RWF) reinforced recycled polypropylene(rPP) composites caused by hygrothermal ageing. RWF is hydrophilic, absorbs water, and the rPP properties are altered when exposed to high temperatures. Moisture causes the wood polymer composite(WPC) interface to degrade, decreasing the flexural properties. The material with a composition of 37.5wt.% RWF(reinforcement), 50.3wt.% rPP(matrix) and 12.2wt.% additives through hot-pressed and then machined according to ASTM D790. Then, Hygrothermal ageing performs on the specimen for 30 days at 50°C and 90% relative humidity(RH) for the first condition and in water immersion at 50°C for the second condition. The water absorption behaviour result follows the Fickian behaviour. The water absorption rate in water immersion(0.96 m^2/s) is higher than exposure at 90%RH(0.95 m^2/s). The thickness swelling increase indicates poor dimension stability of composite in a moisture-rich environment. The initial strength of the composite is 30.49 Mpa. It reduces after exposure to vapour(27.85 Mpa) and further after exposure to water immersion(26.61 Mpa), same with the modulus pattern indicating the deleterious effect of moisture on the flexural strength and modulus of the composite. The strain at the break of the composite increases after exposure, showing the composite becomes ductile due to plasticising effect of the rPP. The unaged sample observed failed due to fibre and matrix fractured, causing a brittle fracture upon exposure to the vapour in 90%RH and the water immersion, the presence of the pull-out fibre with the addition of microcracking and a gap between the fibre and matrix. It indicates that the fibre and matrix interface weaken. It was concluded that hygrothermal ageing changes the composite dimension and reduces the flexural properties with SEM images to support the effect caused by hygrothermal ageing.

DEDICATION

Special dedication to my parents, Mr. Ajang Bit and Mrs. Rosemary ak. Anthony for their moral support and understanding throughout the semester in completing this research study. I would like to thank my family, my siblings, friends, and my lecturers for helping me out,

Thank you, everyone.

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Firstly, I would like to give grace to my Heavenly Father through the guidance of His Holy Spirit so that I can finish my final year report. It is not by my strengthand knowledge that I can go through this, but by His power, love, and wisdom strengthen me.

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Last but not least, to my friend and family, who always motivates me to complete this report. Thank you, everyone.

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LIST OF ABBREVIATIONS

ASTM - American Society for Testing and Materials

CC - Calcium Carbonate

DSC - Differential Scanning Calorimetry

HDPE - High-Density Polyethylene

LDPE - Low-Density Polyethylene

MAPP - Maleic anhydride-grafted polypropylene

MFA - Microfibril Angle

PP - Polypropylene

PVC - Polyvinyl Chloride

RH - Relative Humidity

rPP - Recycled Polypropylene

RWF - Rubberwood Flour

SEM - Scanning Electron Microscope

Tc - Crystallisation Temperature

Tg - Glass Transition Temperature

Tm - Melting Temperature

UV - Ultraviolet

WPC - Wood Polymer Composite

LIST OF SYMBOLS

°C - Degree Celsius

Mm - Millimetre

P - Primary cell wall

S1 - Outer layer of primary cell wall

S2 - Middle layer of primary cell wall

S3 - Inner layer of the secondary cell wall

wt.% - Weight Percentage

CHAPTER 1

INTRODUCTION

In this section, several points are discussed, and these points are divided into six subtopics. The six subtopics are the background of the study, problem statement, objectives of the study, scope, importance of the study, organisation of the report, and this chapter concludes with a summary of chapter 1.

1.1 Background of the study

Wood-plastic composite (WPC) has recently become a dynamic growth material in various industries. Due to their benefits over plastics, such as low density, high specific strength, readiness, and renewability, natural fibres are increasingly used as reinforcement in polymer composites. (Kazayawoko et al., 1999).

Rubberwood flour (RWF) mainly consists of lignin and wood fibres, rich in resources and easy to process. Wood fibres' strength and flexibility and lignin's strength and stiffness make RWF an excellent filler material for WPC. While minimising environmental problems, permit the use of wood waste as a source of RWF. Recycled polypropylene (rPP) is a standard engineering thermoplastic used in WPC. rPP, on the other hand, has low strength and modulus. Any recycled plastic that can be melted and processed below wood degradation temperatures or other lignocellulose fillers is typically suitable for WPC manufacturing (Clemons, 2008).

When exposed to moisture or submerged in water, RWF or rPP absorbs it. (Dhakal et al., 2007). Unfortunately, the material degrades due to the impacts of moisture absorption, particularly in composite interfaces. Reduced mechanical characteristics of the composite are caused by damage to the interface of the RWF reinforced rPP composite. Therefore, research on how WPC absorbs moisture and how moisture affects mechanical qualities is essential. There have been numerous investigations on the water absorption of natural fibre reinforced polymer composites. It has been confirmed that water molecules have a degradative effect on composite, and absorbed water impacts the materials' mechanical properties. (Perrier et al., 2017). It is well known that fibre or polymer modification can enhance the mechanical properties of natural fibre reinforced polymer composites. (Cai et al., 2016).

In this research, RWF reinforced rPP in WPC is a desirable solution for environmental issues due to the industries' significant amount of wood waste. Both materials are low maintenance and inexpensive material for construction and manufacturers. Preventing wood waste significantly increases wood use efficiency, helps reduce environmental impact, and meets the demand for wood products without damaging environmental resources.

1.2 Problem statement

Wood is one of the most commonly utilised materials in the construction sector. The industry's demand for wood products is expanding rapidly, necessitating using landfills to dump trash and creating environmental problems. Leftovers such as excess pile products from wood need to dump appropriately. In addition to wood, polypropylene waste will also cause damage to the environment since it needs a longer time to decompose. It must be recycled to ensure raw materials are not wasted or dumped in landfills.

RWF and rPP can be used to create WPCs for construction materials. However, the most significant drawback of natural fibre-reinforced thermoplastic composites is their propensity to absorb moisture when submerged in water or exposed to humid conditions. (Dhakal et al., 2007). RWF nature is hydrophilic. It absorbs water from the environment, and when ageing in hydrothermal conditions causes its properties to degrade. Thermoplastic material when exposed to temperature, primarily when used above glass temperature (Tg). They may undergo thermal degradation (Bazli and Abolfazli, 2020). In addition, rPP is hydrophobic, but the disadvantage of rPP is that its strength and modulus are relatively low under high thermal conditions. Moisture absorption in WPC from RWF reinforced rPP causes the material to degrade, especially in composite interfaces. Damage to the composite interface affects the stress transfer, decreasing mechanical properties.

This study aims to understand better the characteristics of WPC made of RWF and rPP when subjected to high temperatures and humidity. Investigations were done on the mechanical and thermal characteristics of rPP and RWF composites. When using RWF reinforced rPP composites materials for the construction sector, understanding the nature of RWF reinforced rPP gives one an advantage in resolving issues. Therefore, using RWF and rPP to manufacture WPC improve environmental issues, especially landfills.

1.3 Objective of the study

The objectives of the study are stated as below:

1. To compare the effect of hygrothermal ageing on the physical properties of rubberwood reinforced recycled polypropylene composites.
2. To compare the effect of hygrothermal ageing on the flexural properties of rubberwood reinforced recycled polypropylene composites.
3. To evaluate the fracture surface of rubberwood reinforced recycled polypropylene composites caused by hygrothermal ageing.

1.4 Scope

The material used in this study is rubberwood reinforced recycled polypropylene composites in the form of a granule consisting of a weight percentage(wt.%) of 37.5% rubberwood flour, 50.3% recycled polypropylene, 3.9% maleic anhydride grafted polypropylene, 0.2% ultraviolet, 1% lubricant, and 7% calcium carbonate supplied by the Prince of Songkla University, Thailand. The specimen was shaped using a hot compression moulding process and then machine according to ASTM D790-92 standards. The specimen then aged under high temperatures. High relative humidity (50°C and 90% RH) in the environmental chamber above glass transition (Tg) for polypropylene (11.63°C) and some WPC samples aged in water immersion at 50°C for 30 days. The weight changes were monitored at the selected interval period. The unaged and aged specimen characterised in the flexural test using a Universal Testing Machine and fracture surface behaviours using Scanning Electron Microscope (SEM).

1.5 Significant of the study

This study may contribute to the construction industry by using new materials used in construction. By implementing a WPC design based on RWF reinforced rPP, it will be able to create low-cost materials and, at the same time, help the environment.

WPCs granule made of RWF reinforced rPP composites compressed and moulded using a hot press. Understanding its features provides the knowledge to overcome weaknesses and makes the WPC manufacturing process more straightforward. Using RWF reinforced rPP reduces wood waste, significantly improves the efficiency of using wood, and helps reduce landfills that can impact the environment. The consumption of virgin plastics and the environmental impact is decreased by recycling polymeric materials after use. The majority of petroleum-based single polymer plastics are relatively simple to recycle.

So, unwanted plastics can be recycled into new items with just the addition of energy using an effective collecting, sorting, and recycling system.

Finally, by conducting this study and understanding its character, RWF reinforced rPP can be enhanced suitably for the construction industry. At the same time, the landfill number to dispose of wood and polypropylene waste is reduced, saving the environment from pollution and using more natural resources.

1.6 Organisation report

Chapter 1 introduces the study "Hygrothermal ageing for recycled polypropylene and rubberwood waste composites for construction materials". This chapter contains the background of the study, problem statement, objectives of the study, scope, organisation of the report and summary.

Chapter 2 reviews past studies on "Hygrothermal ageing of recycled polypropylene and rubberwood waste composites for construction materials". Reviews related to wood polymer composites also be covered. This chapter also discusses composite materials based on history, classification, main properties, applications and fabrication processes.

Chapter 3 explain the method and material used to study the "Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials" for construction materials. This research analyses the water absorption, morphology, and physical and flexural properties of recycled polypropylene (rPP) reinforced rubberwood flour (RWF) composites. This methodology contains material, compression moulding method, humidity, water immersion, flexural testing, and morphological testing.

In chapter 4, after the experiment test has been done. The result shows altogether with the discussion regarding the "Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials" to achieve the objective of this project.

Finally, in chapter 5, the conclusion of the results of the “Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials” are revealed. This chapter includes recommendations and suggestions for further research.

1.7 Summary

This research introduces the study of “Hygrothermal ageing for recycled polypropylene and rubberwood waste composites for construction materials”. Chapter 1 contains the background of the study, problem statement, objectives of the study, scope, and the importance of the study, and organisation of the report. The summary of this chapter shows that environmental issues should be reduced by manufacturing WPCs from RWF-reinforced rPP. However, the character of RWF and rPP has its drawbacks. Then, a study on the thermal and mechanical properties of RWF reinforced RPP must be done to overcome the problem. Chapter 2 reviews past studies related to this research and reviews related to wood polymer composites. This chapter also discusses composite materials based on history, classification, main properties, applications and fabrication processes. Chapter 3 explain the method and material used during this study. This research analyses the properties of rPP reinforced RWF composites. This methodology contains material, compression moulding method, humidity, water immersion, flexural testing, and morphological testing. Chapter 4, after the experiment test, has been done. The result shows altogether with the discussion regarding the study that has been done to achieve the objective of this project. Finally, in chapter 5, the conclusion of the results of the “Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials” are revealed. This chapter also includes recommendations and suggestions for further research.

CHAPTER 2

LITERATURE REVIEW

This chapter reviews past studies on "Hygrothermal ageing of recycled polypropylene and rubberwood waste composites for construction materials". Reviews related to wood polymer composites are also covered. This chapter also discusses composite materials based on history, classification, main properties, applications and fabrication processes.

2.1 Wood polymer composite

Composites are hybrid materials made of fibre-reinforced polymer resins, combining high mechanical and physical performance fibres and the physical properties of polymers. A composite material is created when two or more different materials are joined to make a structural component. The composite is made of two materials: one serves as a continuous matrix, and the other serves as reinforcement or filler. Engineering composite materials is essential because it creates a material by combining two or more separate elements that have superior or crucial properties over the properties of the individual components (Dutt et al., 2020).

Thus, composite materials are formed by aligning solid and rigid constituents such as fibres and particles in a matrix binder. Materials in this class have high mechanical properties. One of its components accommodates the stress to combine components called the reinforcement phase. Another provides a strong bond called the matrix. Polymers, ceramics and metals have found applications as matrix materials. The reinforcement phase is another component in a composite called reinforcement and can be fibres, particles or laminates. Different components and their interactions' compatibility affect the composites' properties. An extensive study has been done to pinpoint the variables influencing the mechanical behaviour of composite. As particle size decreases and matrix adhesion increases, the reinforcing effect often rises.

Composite materials are divided into categories depending on the continuous and discontinuous phases (Zhandarov and Mäder, 2005). Discontinuous reinforcement phases, such as fibres and fillers, are classified as fibre and particle composite. Reinforcement is a strong, inert nonwoven fibre material placed into a matrix to enhance the physical qualities of the metal glass. The dimensions of the reinforcement determine the capabilities that contribute properties to the composite. Fibres are very effective in increasing fracture resistance because reinforcements with long dimensions do not promote early fracture growth that is common to reinforcement which can otherwise lead to failure, especially with brittle matrices. Figure 2.1 shows the classification of natural fibres used in polymer composites.

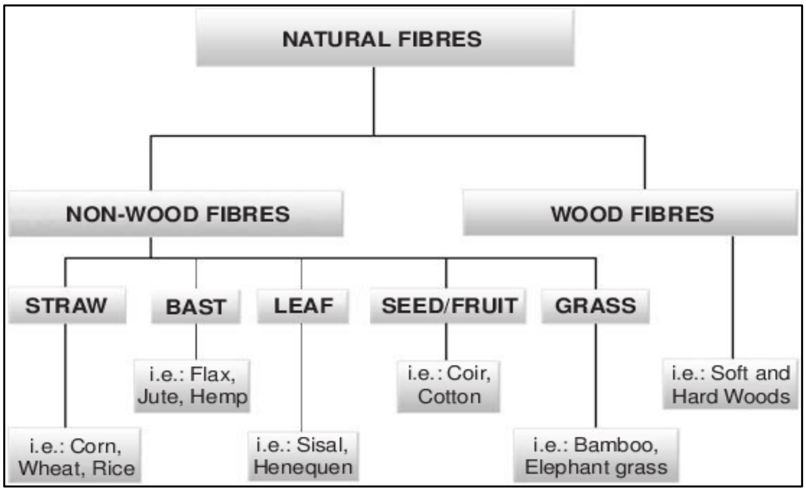


Figure 2. 1 Natural fibre classification used in polymer composites (Puglia et al., 2005)

Wood flour and thermoplastic resins like polypropylene, polyethene, or polyvinyl chloride are the main ingredients in wood-plastic composites. Kim and Pal (2011) reported that WPCs could be produced using environmentally benign materials, including discarded wood, recycled thermoplastics, and underutilised natural resources. High durability, wear resistance, incredible strength, and superior stiffness are some of WPC's fantastic qualities. High moulding performance and a texture like genuine wood are different characteristics. Making outdoor decks for the construction industry is another use for WPC. One of the factors affecting the mechanical characteristics of WPC is wood flour. The incompatibility between hydrophobic thermoplastic resins and hydrophilic wood flour must be addressed to improve the mechanical properties of WPC. Wood flour has undergone chemical and physical modifications to increase its compatibility with thermoplastic resins, including corona, plasma, and ionising radiation. (Wei et al., 2013).

2.2 Wood fibres as reinforcement

For many years, wood has been utilised to strengthen the fillers in thermostat polymers. However, the use of wood fibre in thermoplastics is comparatively new, thanks to advancements in processing and coupling agents. Compared to inorganic fillers, using rubberwood as a filler in WPC offers benefits such as biodegradability, low cost, renewable, low specific gravity, and less equipment damage. The physical properties, chemical composition, and microstructure depend on the wood species, such as hardwood and softwood. Therefore, the wood species selection in WPC can significantly affect the microstructure and properties.

Rubberwood may be in wood flour, sawdust, wood powder, wood fibres, or eight pulp available for WPC production. However, rubberwood should be ground into fine flour or refined into fibres for plastic-based composites. When using rubberwood flour, the reinforcement of wood flour on plastic is not by using individual fibres but particles consisting of bundles of broken wood fibres. Tajvidi and Azad (2009) found that extruded samples showed an increase in young modulus with wood flour particle size in 180–425µm.

The smallest particle size is found with the lowest water absorption and thickness. Stark (2001) has reported that composites made of wood flour and polypropylene with various wood particle sizes show aspect ratios, not particle sizes, having the most significant effect on strength and stiffness.

The use of rubberwood fibres has little effect on the impact energy. However, in the case of rubberwood fibres, the tensile strength and young's modulus are reduced. The failure stress increases by reducing the composite's average fibre length, with the wood fibre field ratio between 16 and 26. This higher aspect ratio increases the stress transfer from the matrix to the fibres. Many commercially made wood powders used as thermoplastic fillers are less than 0.425mm, with an aspect ratio of 3.4 (Macromolecules, 2005).

In comparison to coarser wood flour, very fine wood flour costs more and increases the viscosity of the melt. Composites typically develop a smoother finish and a more uniform look, though. If the fibre bundles are ground too finely, they turn into wood dust and stop looking like fibres or fibre bundles.

2.2.1 Structure of woods

Hardwood and softwood are two types of woods (Joffre, 2014). Rubberwood is in the softwood fibre classification. Hardwood fibres are generally shorter, about 1 mm, and less flexible. In comparison, softwood fibres are longer, between the 2-3 mm range (Tabarsa and Chui, 2001). Wood fibres are usually presented as a hollow layered structure. It is defined as the primary cell wall (P), the outer layer (S1), the middle layer (S2) and the inner layer (S3) of the secondary cell wall. The different layers are illustrated in Figure 2.2.

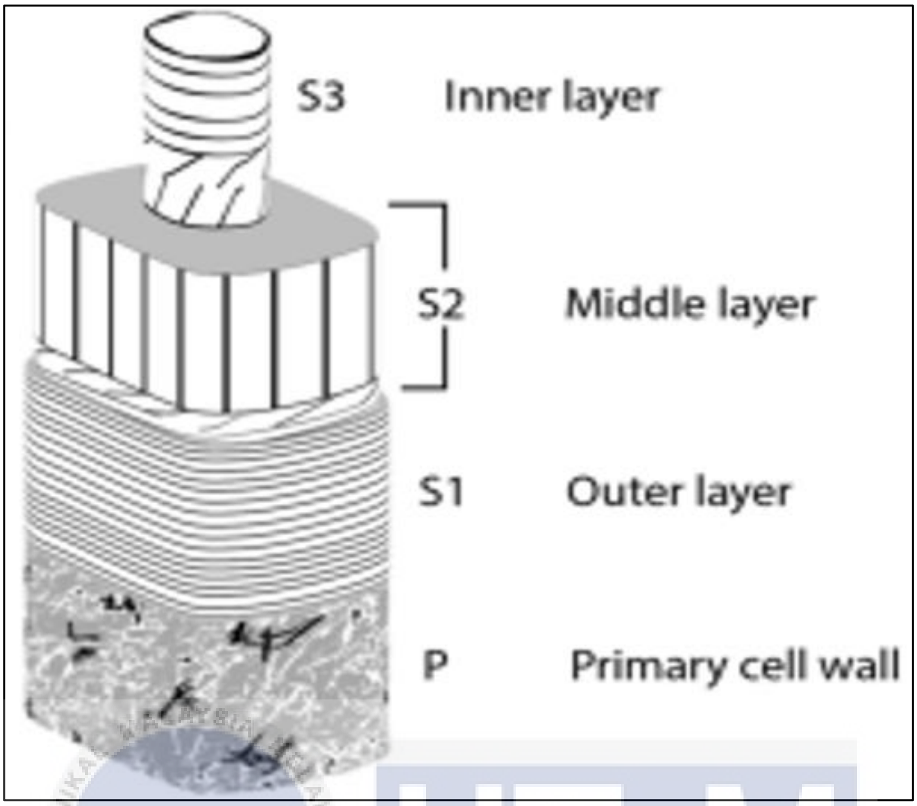


Figure 2. 2 Diagram of the softwood cell wall (Joffre, 2014)

The primary part of the cell wall is formed by the secondary fibre wall (layer S2), affecting the mechanical properties and the physical of the fibre. Each layer consists of several polymers of lignin, hemicellulose and cellulose. Cellulose microfibrils wrap around the fibre axis with an angle known as the microfibril angle (MFA). MFA depends mainly on the direction the tree grows. When a conifer shoot moves from its standing position, the bottom of the shoot forms a compression stick. Compression wood typically has a higher MFA than regular wood (Andersson and Tillman, 1989). The growth rate of compression wood increases the top, resulting in renewed horizontal growth (Yamamoto et al., 2001). Suppose the cylinder approaches the geometry, the late wooden tracheid’s cross-section, a polygon in the green state, a significant deviation. In composites, fibres are extracted from wood. They show different hydro elasticity properties due to mechanical or chemical treatment during the pulping process.

2.2.2 Composition of wood

Wood is categorised as a lignocellulose substance and is composed primarily of the three chemical components cellulose (45–50%), hemicellulose (20–25%), and lignin (20–30%). 0–0.5% of other insignificant ingredients and 1–10% of extractives (Jiang and Kamdem, 2004). The chemical composition of wood is different between types of wood.

2.2.2.1Cellulose

The cellulose is found in the primary structural parts of the wood. An anhydrous glucose molecule with three oxygen, carbon, and hydrogen atoms organised in a straight line and joined by glycosidic bonds makes up the cellulose molecule (Figure 2.3). With a polymerisation rate of about 10,000, it possesses a very high concentration of anhydrous glucose units in a crystalline and linear polymer. Cellulose, which typically contains 60–90% crystals by weight, is the primary factor that provides the wood with its strength and structural stability. The surface hydroxyl groups on cellulose are primarily responsible for its molecular organisation. Hydrogen bonds between molecules are held together by a high proportion of crystalline cellulose. A hydroxyl group may be between glucose units in two adjacent molecules or the same molecule responsible for its reactive properties. Cellulose is hygroscopic because it contains polar molecules and readily undergoes hydrogen bonding (Jiang and Kamdem, 2004). The water absorption by cellulose depends on the number of free hydroxyl groups. Water molecules can reside in the amorphous region but cannot enter the crystal region (Stokke and Gardner, 2003).

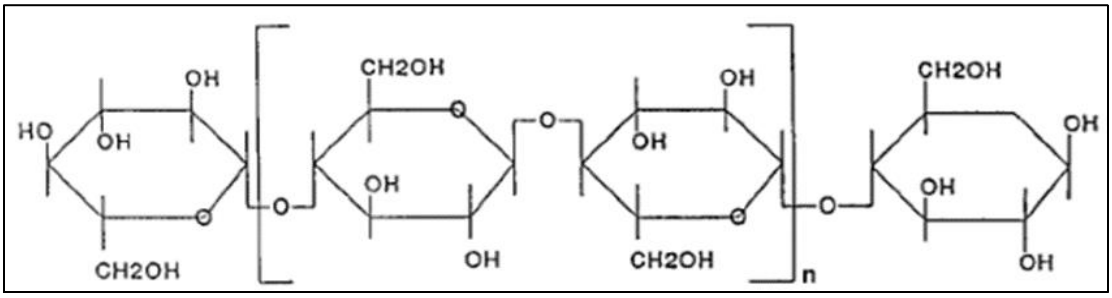


Figure 2. 3 The linear and unbranched structure of a cellulose molecule (Stokke and Gerdner, 2003)

2.2.2.2 Hemicellulose

Hemicellulose comprises a variety of polysaccharides that have less polymerisation than cellulose. Only tens or hundreds of repetitive units are polymerised at this level. The only difference between its structure and that of cellulose is that hemicellulose is structured as 5 or 6 carbon sugar chains. In contrast to cellulose, the chains are less organised and relatively short. Hemicellulose is therefore readily broken down or soluble.

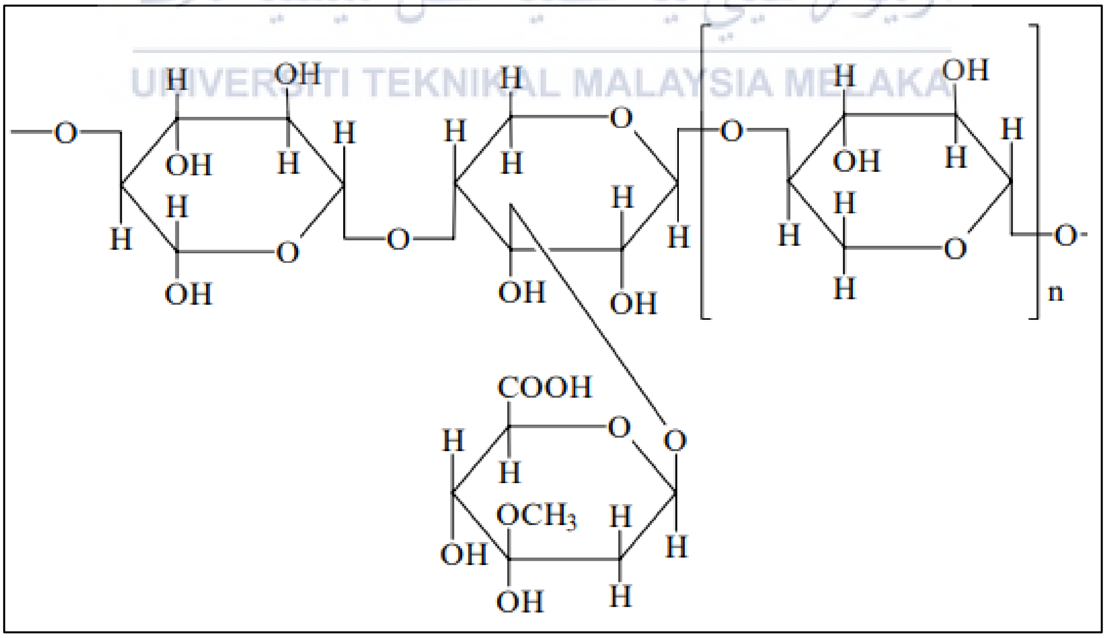


Figure 2. 4 Hemicellulose structure (Alam et al., 2014)

2.2.2.3 Lignin

Lignin is a binding agent that holds cellulose fibres together. It is a largely inactive substance that serves as a binding and hardening agent. It is built into a significant phenyl propane building block, usually containing a hydroxyl group in the meta-position to the side chain (Hafezur et al., 2013). Through the diffusion of lignin into the fibre wall, the stiffness of the wood cell enables stress transfer between the matrix and the filler in enhanced WPC. It is made up of oxygen, carbon and hydrogen. Lignin is inactive compared to cellulose due to low hydroxyl sites. In addition, carbon to oxygen or carbon to carbon may be attached to an aromatic ring on the structural part. Due to the alcohol and phenolic hydroxyl groups, polyfunctional lignin can exist with two or more neighbouring chain molecules. Postulated monomers in lignin are shown in Figure 2.5.

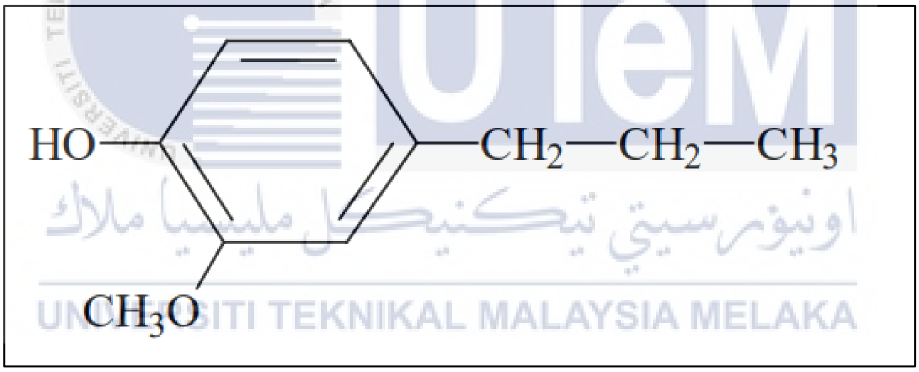


Figure 2. 5 Lignin molecule structural unit (Alam et al., 2004)

2.2.3 Properties of woods

The mechanical properties of wood are excellent in terms of tensile, compression, shear, hardness, static bending and impact bending. Each test determines the stresses per unit load area and strength between criteria, such as modulus of elasticity, flexural strength and toughness. Laboratory data were analysed to create the stress data values utilised by engineers and architects when developing wooden structures. Tracheids and fibres are also put through testing because their tensile strength is correlated with the product's tensile strength.

The strength index of clear wood can best be predicted by density, with higher densities suggesting stronger wood. When it changes regularly at the fibre saturation point, moisture content also impacts wood's strength. Most strength qualities increase when the moisture content is reduced (Tsoumis, 2020). Strength is also impacted by temperature and loading time. Generally speaking, strength declines as temperature rises. Compared to short-term laboratory experiments, permanently loaded wood can withstand a lower maximum load.

Although wood expands and contracts with changes in temperature due to its thermal characteristics, the shrinkage and swelling brought on by variations in moisture content are much more significant. Temperature-related expansion and contraction are typically ignored and have little real-world application. Compared to metal, concrete, glass, and marble, wood has low thermal conductivity. The thermal conductivity rises with moisture content and density and is most significant in the axial direction. Wood burns when exposed to hot enough temperatures. These characteristics make wood advantageous for warmth but disadvantageous for technological usage. Compared to hardwood, softwood has a higher heating value and extractive impact. Air-dried wood has around 15% less heating value than oven-dried wood because humidity lowers the heating value.

2.3 Polymer in WPC

Both thermoset and thermoplastics can be used in the production of WPC. Thermoplastics are based on slightly branched, or linear, polymers chains of molecules that flow with each other. Plastics are in solid form at room temperatures and deformed at high temperatures, and their process can be reversible. It can undergo many solidifications and melting cycles without significant chemical changes, making it ideal for recycling.

The polymer matrix can be thermoplastic or thermoset. Thermoplastic polymers are branched or linear molecules that are soft and melt when heated. Thermoset-based composites are cheaper than thermoplastic-based composites but have weaker toughness when tested in interlaminar mode and lower thermal distortion temperatures. Thermoset polymers include epoxy, phenolic resins and unsaturated polyesters. At the same time, thermoplastic polymers are saturated polyesters, polycarbonates and polyamides (Wolf, 2000). Many thermoplastic-based composites provide excellent impact resistance and are ideal for high-performance applications.

Thermoset is a polymer network formed by cross-linking reactions and cannot be remelted again. Thermoplastics are frequently employed as matrix materials for natural fibre composites, mainly wood. The processing temperature of thermoplastics in composites should be lower than the temperature at which wood thermally degrades. Andersson and Tillman (1989) found that due to the limited thermal stability of wood, thermoplastics that meet this requirement, including LDPE, HDPE, PP, and PVC, are suitable for use in WPC either in recycled or virgin forms. However, PVC was the first thermoplastic used commercially in WPC manufacture. However, PE is now the most widely used type today, followed by PP for WPC among all thermoplastics (Kozlowski, 2002). Polypropylene is stiffer, more chemical and scratch-resistant while still very hard.

2.3.1 Polypropylene

Polypropylene is the polyolefin that is most frequently utilised to create composite materials. Following its controlled polymerisation, this commodity polymer has been produced commercially. Polypropylene can be blended with a wide range of various fillers or reinforcing agents. Many additives are being developed to enhance polypropylene’s thermal resilience to minimise degradation. The absence of waste catalysts and impurities is another essential condition for polypropylene to be used in the production of composites. Breakthroughs in polypropylene and polyethene copolymers have considerably impacted the use of low-cost, rigid thermoplastic composites.

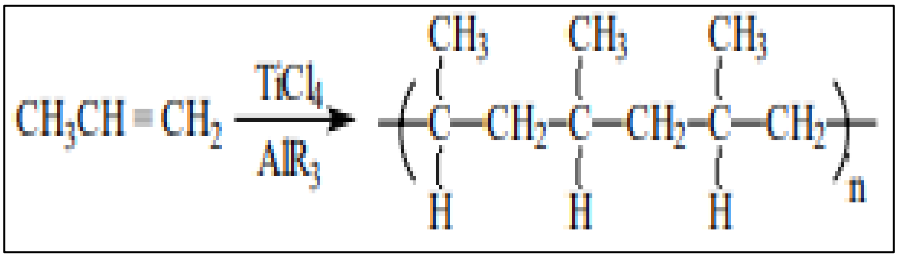


Figure 2. 6 Stereoregular (Hossain, 2013)

2.3.2 Polypropylene molecular structure

The molecular structure of various types of PP and the introduction of universally used descriptive nomenclature today were first determined by Natta in 1955 (Bismarck et al., 2002). The commercial benefit lies mainly in the highly crystalline PP and further modification through copolymerisation. In isotactic polypropylene, each monomer unit is organised straight from head to tail without branching. Each methyl group has the same structure, as seen in the figure below. Such a stable structure is a suitable catalytic characteristic. It is due to the type of template that constrains the stereospecific catalyst site. Methyl groups alternately occupy the carbon chain in the syndiotactic form. Damage of the kind depicted in Figure 2.7 C results from defective monomer insertion. Figure 2.7 B illustrates a severe fault in atactic polypropylene that finally results in the loss of steric control. Although theoretical, syndiotactic polymers have little commercial significance (Whiteley, 1992).

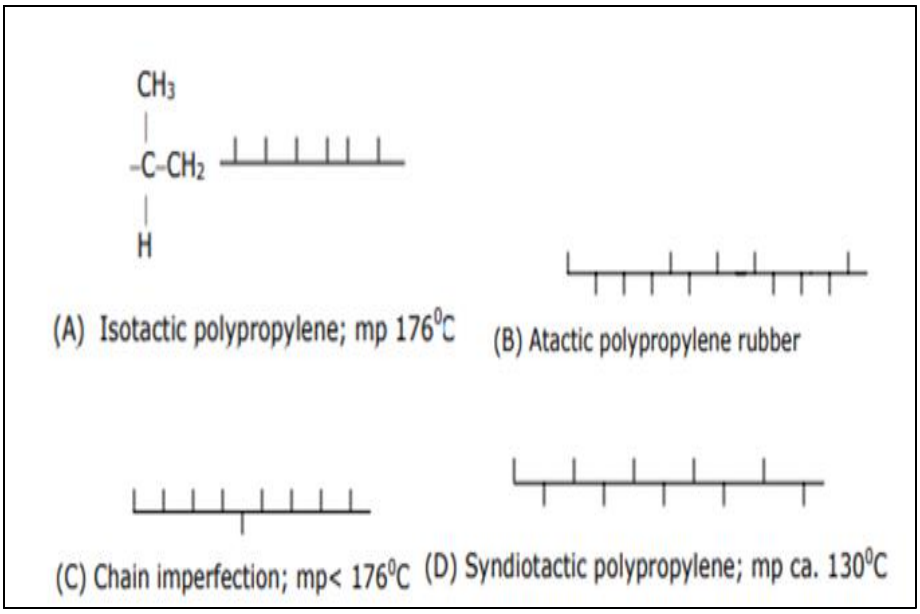


Figure 2. 7 Chain structure of polypropylene (Hossain, 2013)

2.3.3 Polypropylene crystallisation

Polypropylene specialities come from its ability to crystallise and form stiff and rigid particles. Polypropylene is considered a semi-crystalline polymer (Whiteley, 1992). It has two constant crystalline and amorphous phases. The highly isotactic polymer forms a helical coil with a stable structure with three-unit of monomers per round. These coils overlap into thin lamellar crystals, forcing the chain to be folded several times as it emerges and re-enters the lamellae (Whiteley, 1992). Three crystal forms are known, the α form being the most stable, preferred at higher temperatures and slower cooling rates. Rapid quenching produces a β-form with a lower density and melting point of 150°C. Random copolymers and lower stereoregularity polymers often include a low-melting -crystal in addition to the α form.

The crystallite formation from the viscous melt is obstructed by chain entanglement and the need for helices. If it closes the pack, fold the pack into a lamella. The formation of nuclei in the polymers contributes to the low crystallisation levels, even for highly stereoregular polymers. Commercial particles vary from 30% crystallinity in films quickly extinguished to 50-60% in moulds. Even stereoregular polymer samples that have been purified and annealed barely reach a crystallinity above 70%. It should be emphasised that the high helix content is not synchronised with the high crystallinity, which requires a three-dimensional long-distance arrangement. The crystal properties of PP are given in Table 2.2. The dominant and stable crystal forms have a monoclinic structure. More stable forms are preferred at higher temperatures and slower cooling rates.

Table 2. 1 Crystalline properties of Polypropylene (Hossain, 2013)

Crystal form	System	Crystal density (200C), g/cm^3	Chain per unit cell	MP, °C
so α	Monoclinic	0.932-0.943	4	171
so β	pseudo-Hexagonal	0.922	9	150
so γ	Triclinic	0.939	1	131
Amorphous	-	0.85	-	-
Syndio	orthorhombic	0.93	2 (4/1 helix)	138

2.3.4 Properties of Polypropylene

Polypropylene is a linear hydrocarbon polymer that contains zero unsaturation or little saturation. Therefore, polypropylene and polyethene have many properties and similarities. The fundamental mechanical properties are influenced by molecule mass distribution, molecular mass, chain stereoregularity, and processing conditions that add structural orientation and strain. Several additive packages must be synchronised by the application to ensure excellent performance. Some properties of polypropylene are given in Table 2.3. The final melting point of commercial polypropylene is 160-170°C, where the pure polymer around reaches 176°C. The higher the amount of isotactic material, the crystallinity of the polymer will be more excellent. The crystalline and non-polar properties of polypropylene provide outstanding resistance to various polar and aqueous media, including emulsified solutions with intense stress cracking abilities. The disadvantage of polypropylene is that the methyl group is susceptible to thermo-oxidative degradation.

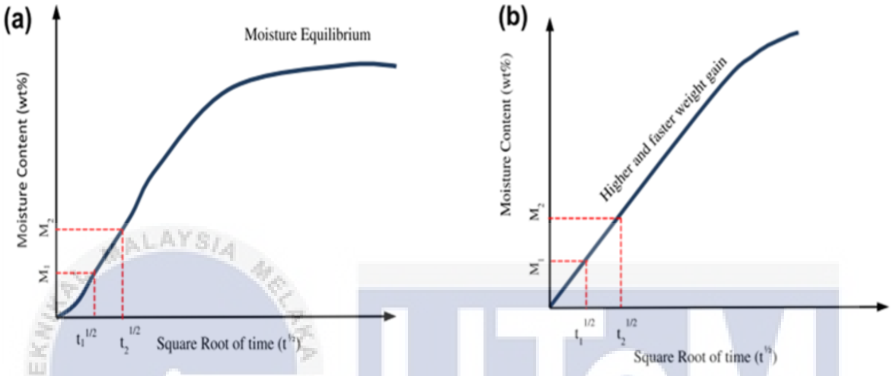
Table 2. 2 Properties of Polypropylene (Hossain, 2013)

Parameters	Standard Used	Values
Specific gravity	ASTM D792	0.90-0.91
Crystallinity, %	-	82
Melting temperature, °C	-	165-171
Tensile strength, MPa	ASTM D638	31-41
Tensile modulus, GPa	ASTM D638	1.10-1.55
Elongation-to-break, %	ASTM D638	100-600
Impact strength notched Izod, Jm-1	ASTM D256	21-53
Rockwell hardness (R-scale)	ASTM D785	90-95
Heat-deflection temperature, °C, at 455 kPa (66psi)	ASTM D648	225-250

2.4 Hygrothermal Ageing in WPC

WPC is made from inexpensive, lightweight wood fillers that may be processed using thermoplastic polymers. WPC has been utilised in outdoor applications since it first entered the market. The market share of WPC decks has proliferated from 2% in 1997 to 18% in 2005 (Smith and Wolcott, 2006). WPC is known for its low maintenance and high durability compared to pressure-treated wood. WPCs used in outdoor applications are exposed to areas where extreme humidity and temperatures damage them. However, the humidity and temperature of the environment affect the WPC.

The pattern of desorption and absorption of water at room temperature is followed by Fickian behaviour. The water uptake process starts linear, slows down and eventually reaches saturation. Water diffuses due to concentration gradients from places of the highest concentration to the lowest, a process is known as Fickian diffusion. (Bao et al., 2001). The moisture uptake behaviour accelerates higher immersion temperatures, and the moisture saturation time is significantly shortened. This non-Fickian behaviour may indicate differences in water molecule absorption behaviour and state inherent in composites (Dhakal et al., 2007). The diffusion coefficient characterises the capacity of solvent molecules to flow across polymer segments. The composite surface exfoliates and dissolves when the temperature rises in a humid environment, which raises the permeability coefficient. Microcracks form on the surface, and most materials expand as a result. (Joseph et al., 2002). Figure 2.8 shows the graph for Fickian and non-Fickian diffusion, respectively.



a) Fickian diffusion at room temperature

b) Non-Fickian diffusion at elevated temperature

Figure 2. 8 Graphs of Fickian and non-Fickian diffusion (Dhakal et al., 2007)

2.4.1 Moisture transport mechanism

In polymer composites, moisture transport can be facilitated by three mechanisms: diffusion in the matrix, imperfections in the matrix such as micro spaces, pores, or cracks or capillaries along the fibres and matrix interfaces (Beg and Pickering, 2008). The studied composites began to show conductivity after absorbing about 50% of maximum moisture.

Water absorbed in polymers consists of free water and bound water (Chen et al., 2009). While bound water is a dispersed molecule bonded to a polar group of a polymer, free water can flow freely through spaces as more water is absorbed, bound water increases and free water declines. Figure 2.9 shows the moisture conditions in the polymer matrix. When WPCs are exposed to moisture, water penetrates and adheres to the hydrophilic groups of the fibres, creating hydrogen bonds between the molecules and the fibres and reducing the adhesion of the fibre-matrix interface. The deterioration process starts when the cellulose fibres’ swelling places pressure on the interface region, causing a micro-fracture mechanism in the matrix around the swollen fibres that encourages capillaries and transport through micro-fractures. According to research on flax fibres reinforced with unsaturated polyester composites, water-soluble substances start to enter the fibres and eventually cause final dehydration between the fibres and the matrix. (Dhakal et al., 2007).

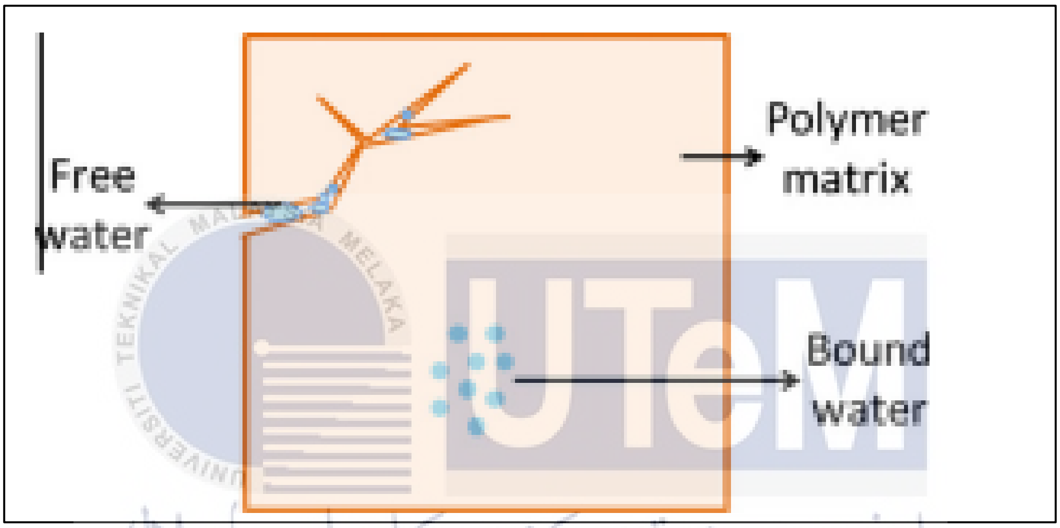


Figure 2. 9 Free water and bound water in a polymer matrix (Azwa et al., 2013)

After an extended period, biological activities such as fungal growth degrade natural fibres (Chen et al., 2009). By creating osmotic pressure pockets on the fibre surface due to the leaching of water-soluble material from the fibre surface, debonding between the fibre and matrix is first caused. (Joseph et al., 2002). This process is summarised in Figure 2.10. The properties of natural fibre composites immersed in water are influenced by the properties of the fibre material and the matrix. Relative moisture and manufacturing techniques determine fibre composite's porosity and volume fraction (Dhakal et al., 2007).

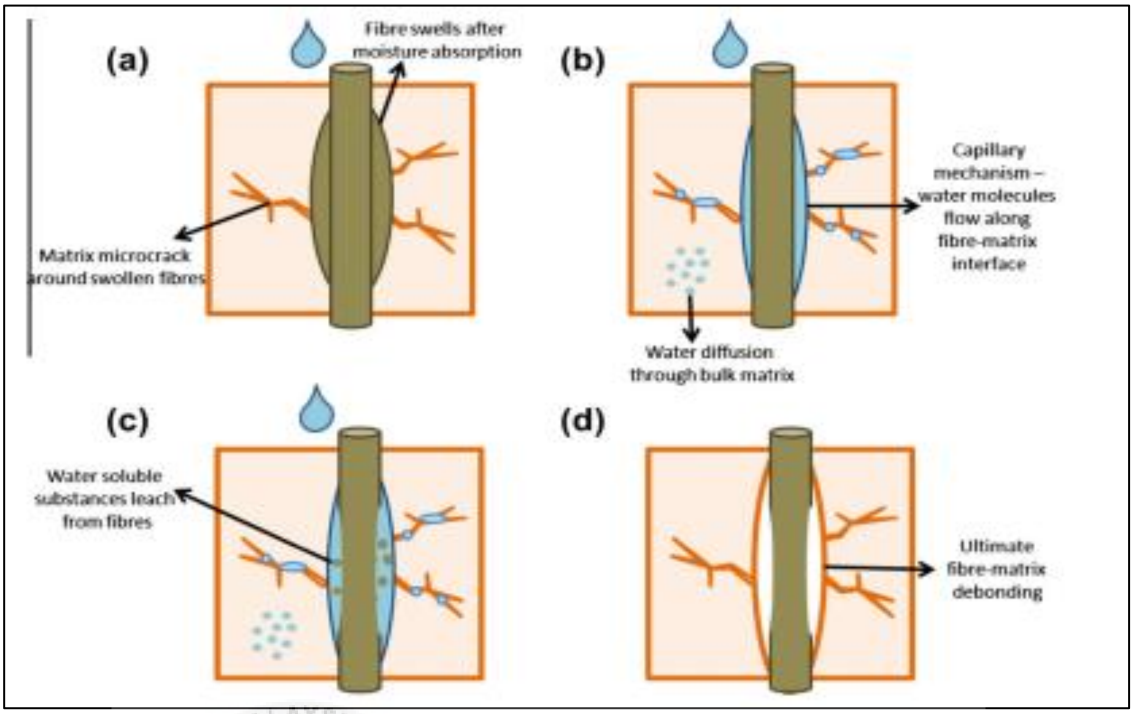


Figure 2. 10 Effect of water on fibre matrix interface (Azwa et al., 2013)

Temperature, fibre volume percentage, reinforcement orientation, fibre permeability qualities, exposed surface area, diffusion, water-matrix reactivity, and surface protection affect how composite materials absorb water. (Joseph et al., 2002). It can be concluded that WPC is sensitive to moisture at high temperatures. Damage caused by moisture is known to accelerate; therefore, additional protection is vital for composites exposed to this condition. Over time, exposure to moisture causes them to lose their function. This aspect must be considered in producing composites that can be used in high humidity environments. One of the suitable characteristics is the selection of suitable fibres to ensure that they have properties suitable for high moisture resistance.

2.4.3 Effect of Moisture on the Properties of WPC

One of the main problems for WPC is its high-water absorption. Cellulose fibres are hydrophilic and absorb moisture (Nabi and Jog, 1999). The moisture content of the fibres can vary between 5 and 10%, leading to dimensional variations in the composite and affecting the mechanical properties of the composite. Much care must be taken during the processing of thermoplastic composites as moisture can cause poor processability and porous products. Pre-treatment fibres with chemicals, or vinyl monomer grafting, can reduce moisture absorption. It has been shown that the moisture uptake of polyolefin-based composites increases linearly with the fibre content. WPC at 20% wood loading absorbs only 1.4% moisture, while 40% loading absorbs up to 9%. The high moisture content reduces the mechanical properties of the composite, especially the bending properties (Stark, 2001).

Natural fibres have higher hydrophilicity than synthetic fibres, allowing for more significant water adsorption, which could speed up the deterioration of polymer composites. One of the main issues that need to be solved for outdoor applications where moisture exposure is common is the mechanical durability of plant fibre reinforced composites. Therefore, understanding the mechanical behaviour and mechanism of hydrothermal ageing is essential for a composite's long-term useful applications. Numerous research has examined the susceptibility of natural fibre composites to moisture uptake using accelerated ageing tests, emphasising how moisture uptake impacts the mechanical and thermal properties of the composite. Scida et al., (2013) studied the influence of hydrothermal ageing on damage to flax reinforced epoxy composites. They discovered that moisture altered the composite's Young's modulus and tensile strength. The ageing duration and temperature were critical parameters.

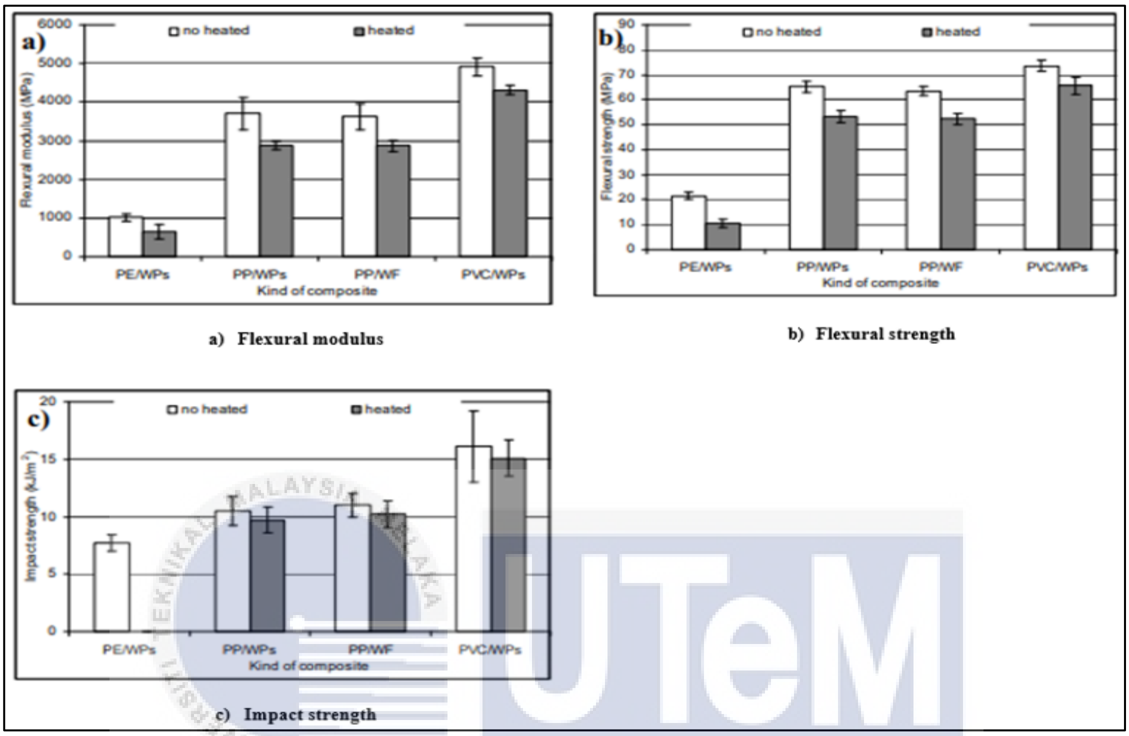
Dhakal et al., (2007) studied the hydrothermal ageing of hemp fibre reinforced unsaturated polyester composites. At higher temperatures, the composites' moisture uptake increased with the fibre volume fraction, resulting in a considerable loss of mechanical characteristics. Alamri and Low (2013) investigated the influence of different weight percentages of nano clay additives on water absorption by natural fibre reinforced composites. They discovered that water absorption considerably reduced the composites' flexural strength, flexural modulus, and fracture toughness. The hydrothermal ageing mechanisms of synthetic fibre reinforced composites have also been extensively researched, including swelling, plasticising, cracking, hydrolysing of the matrix, and debonding of the fibre-matrix interface (Mayandi et al., 2020).

Moisture absorption increases as fibre loading increases. George et al., (1998) studied the relationship between the moisture absorption of pineapple-leaf fibre reinforced low-density polyethene composites and the fibre loadings (10%, 20%, and 30% by weight). They discovered that moisture absorption increased practically linearly as the fibre loading increased. Stark (2001) found that wood flour-polypropylene composites containing 20 wt% wood flour reached equilibrium after 1500 hours in a water bath and absorbed only 1.4 per cent moisture. In contrast, composites containing 40 wt% wood flour reached equilibrium after 1200 hours and absorbed around 9.0% moisture. The investigation revealed that the PP matrix's encapsulation of the wood flour prevents it from absorbing moisture and that the degree of encapsulation is more significant for the composite containing 20% wood flour than for the composite containing 40% wood flour.

2.5.4 Effect of Temperature on the Properties of WPC

The increasing popularity of wood-plastic composite (WPCs) necessitates their new applications. WPC is an ideal substitute for solid wood because WPC can be used indoors and outdoors. However, it does not fully benefit from the effects of external factors. Due to contact with heated metal objects or hot water or the short-term effects of heat and temporary heating of products made of WPC may occur. The effects of temperature increase and thermoplastic effect processing methods cause changes in its molecular structure. These changes can lead to changes in the physical and mechanical properties of the final product.

Gozdecki et al., (2013) reviewed and reported their results in Figure 2.13. The higher temperature at WPC significantly reduces the flexural modulus and flexural strength. It does not significantly affect the impact strength. The most detrimental effect on the mechanical properties of WPC with temperature increase for composites made of PE. At the same time, the most negligible impact was observed for composites containing PVC. Regardless of whether WP or WF was used as a filler, the decrease in the mechanical properties of WPC treated with increasing temperature was at a comparable level.



a) Flexural modulus

b) Flexural strength

c) Impact strength

Figure 2. 11 Mechanical properties of composites (Gozdecki et al., 2013)

However, it is possible to improve the thermal stability of the fibres by coating or binding with monomers. They recently showed that it is possible to reinforce higher melting engineering plastics such as Nylon and PPE with natural fibres by controlling viscous shear heating during processing and using reactive solvents (Jana and Prieto, 2002). The addition of natural fibres improves the thermal properties of composites compared to matrix polymers with increasing Tc crystallisation temperature, Tm melting temperature, and thermal deflection temperature reported (Coutinho and Costa, 1999). This property also depends on the degree of adhesion between the fibre and the matrix.

2.6 Fabrication method of WPC

The mixing process of wood filler with polymer and additives must be done correctly to manufacture consistent composites. Thermoplastic composites typically use two steps: plastic with a coupling agent, wood fillers or other additives, and injection moulding, extrusion moulding, or compounded mixture compression moulding to produce panel-type products. Compounding is the process where dispersing fillers and additives and feeding in a molten polymer using a continuous mixture or batch. The compounding process then produces a plastic pallet of compound wood. The compounding process directly influences the properties of the resulting composites (Bledzki et al., 2005).

Mixing of plastics and wood fillers can be done differently depending on the equipment used, such as pre-mixing with polymers, pre-drying the wood, splitting, and inserting the material into the extruder. Compounding reduces the time for extrusion moulding and reduces the possibility of wood degradation. During compounding, moisture is removed from the wood, improving the quality of the composites.

Compression moulding, injection moulding and extrusion are standard manufacturing processes for WPC manufacturing. The processing method and operating conditions influence the composites' morphology and properties. The compounded material can be formed immediately into a product using injection moulding and extrusion. WPC can be produced by hot flat plate pressing through initial quick press closure, side brackets and final cooling under pressure in large-scale production (Wolcott, 2003).

2.7 Application of WPC

Today, significant applications of WPC can be found in the construction industry (Friedrich and Luible, 2016). Construction and building products are the largest markets for WPC, including decks, fences, doors, exterior windows, garden furniture and others. WPC has the advantage of higher durability, low maintenance and more resistance to warping and debris compared to solid woods. Since WPC is likely to have lower mechanical properties than solid wood, it is unsuitable for applications when strength and stiffness are critical. Building and construction products are the largest markets for WPC.

For infrastructure, using marine and railway intersections is the main application of WPC in the infrastructure sector. WPC does not contain toxic preservatives such as chromate copper arsenate that leak into any river and cause environmental problems. WPC can replace solid wood treated with preservatives for marine use. It has excellent potential as high-quality since wood is becoming hard to find due to restrictions on native species and tropical hardwoods. Moreover, WPC used for rail bonding seems suitable because of its stable and durable nature under challenging conditions.

Finally, WPC is also available in the transportation sector for highway and automotive applications. WPC is used as a substrate for interior uses such as door panels, roof liners, seatbacks, light tone covers and luggage liners in cars. Vinyl, carpets, and other coverings applications cover the substrate. For application on the highway, including highway signage, noise barriers, and fence posts.

Figure 2. 12 Application of WPC for outdoor staircase (Segerholm, 2007)

CHAPTER 3

METHODOLOGY

This chapter explains the method and material used to study the “Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials”. This research analyses the water absorption, morphology, and physical and flexural properties of rPP reinforced RWF composites. This methodology contains material, compression moulding method, humidity, water immersion, flexural testing, and morphological testing.

3.1 Project planning flow chart

A project planning flow chart was planned based on the research topic and used as a guide in this research. The project planning summary is illustrated in the flow chart shown in Figure 3.1.

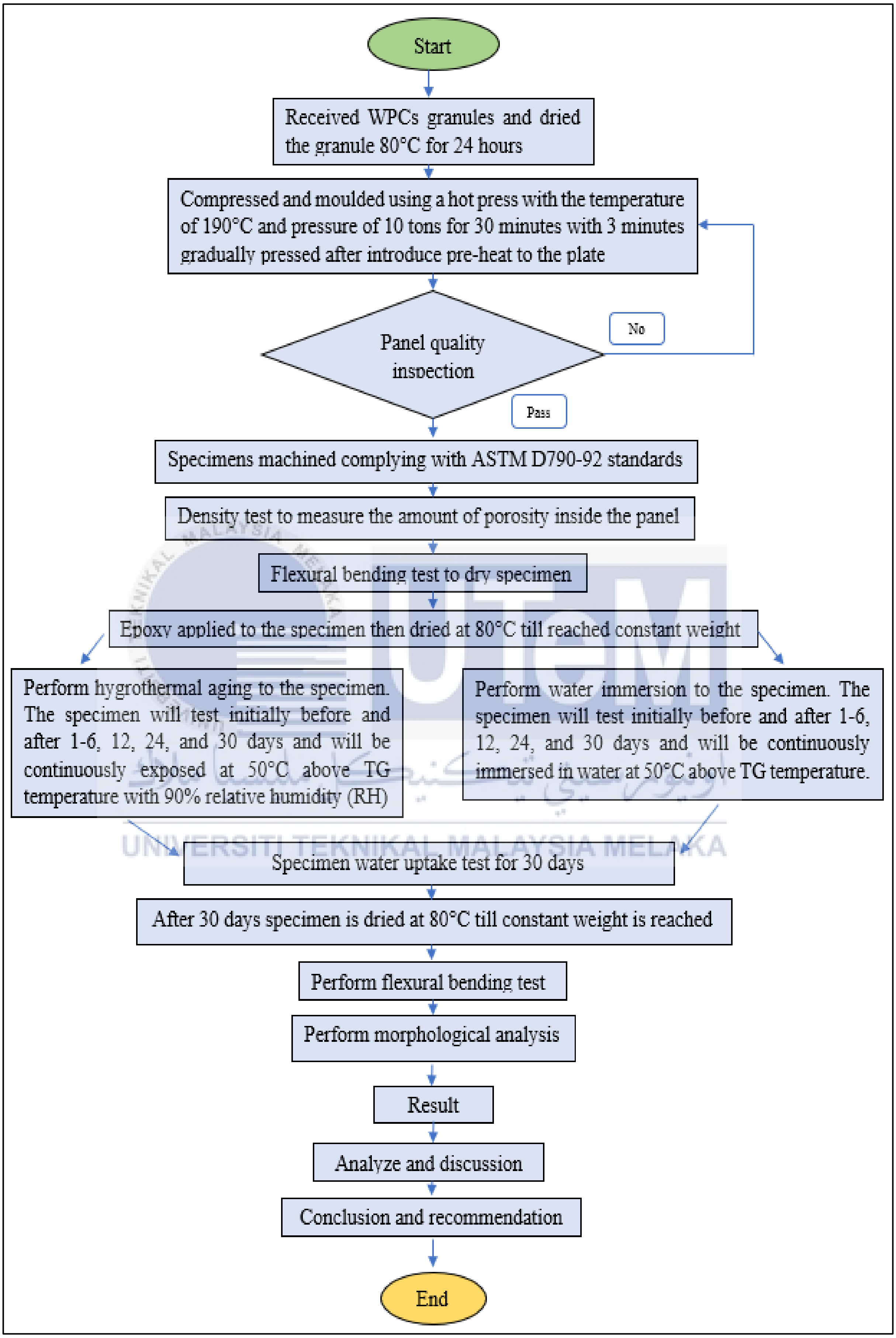


Figure 3. 1 Flow chart of project planning

3.2 Material

The material used in this research in the form of granule rubberwood reinforced recycled polypropylene composites that contain a weight percentage (wt.%) of 37.5% rubberwood flour (RWF), 50.3% recycled polypropylene (rPP), 3.9% Maleic anhydride-grafted polypropylene (MAPP), 0.2% ultraviolet (UV), 1% lubricant, and 7% calcium carbonate (CC). The granule is supplied by the Prince of Songkla University, Thailand.

Figure 3. 2 WPC granule

3.3 Method

This section shows the method used during this study. It includes the material preparation, ageing process conditions and characterisation used in this study.

3.3.1 Compression moulding

The granules were dried in an oven at 80°c for 24 hours to remove the moisture. Then the granule is compressed in a hydraulic hot press machine, as shown in Figure 3.3, having a temperature of 190°C at a pressure of 10 tons for 30 min with the sequence of pre-heating, compressing, and cooling. After the composite panels are made, the panel is machined, complying with ASTM D790-92 standards before any tests are carried out. The specimen was machined according to the specimen dimension shown in Figure 3.4, complying with ASTM D790-92 standards.

Figure 3. 3 Carver 4128 Model 30-12H Manual Heated Press (30 Tons)

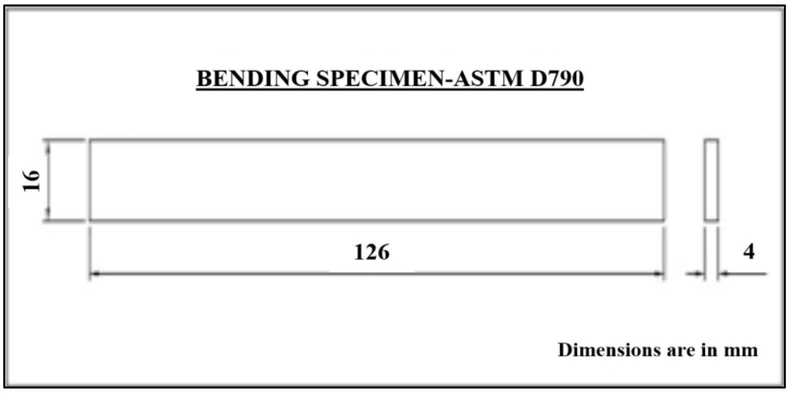


Figure 3. 4 Specimen dimension complying with ASTM D790-92 standards

3.3.2 Hygrothermal ageing

The hygrothermal ageing process performs under two different conditions. The five specimens for each condition dried at 80°C until they reached constant weight before ageing.

3.3.2.1Humidity test

The specimen aged under hygrothermal ageing in high temperature and high humidity. Before the humidity test, the specimen is dried at 80°C in the drying oven (Figure 3.5) until constant weight is reached. All the specimens were exposed under simulated temperature at 50°C, above Tg temperature of 11.63°C for pp and 90% relative humidity (RH). These conditions were simulated using a humidity chamber (Figure 3.6). The specimens through a water absorption test for 30 days of continuous conditioning at 50°C and 90% RH. All the samples will be weight before the test.

Figure 3. 5 GAINSON WGL-65B electric drying oven

Figure 3. 6 Memmert HCP 108 115V HCP Humidity Chamber

3.3.2.2Water immersion

The specimen was immersed in water immersion under high-temperature conditions. Before performing hygrothermal ageing, the specimen is dried at 80°C until constant weight is reached in the drying oven (Figure 3.5). All the specimens were exposed under simulated temperature at 50°C, above Tg temperature 11.63°C for pp. These conditions were simulated using a water bath machine, as shown in Figure 3.7. The specimens through a water absorption test for 30 days of continuous conditioning at 50°C in water. All the samples were weight before the test. After water absorption analysis, the specimen is dried at 80°C until it reaches constant weight to remove excess water in the specimens.

Figure 3. 7 Memmert WNB 7 Water bath

3.3.3 Moisture absorption behaviour

During hygrothermal ageing, the specimen is exposed to water, high relative humidity and high temperature. The specimen absorbs moisture and swells during the ageing process. Moisture absorption analysis is conducted on the specimen for 30 days duration. The percentages of moisture uptake are calculated by using Equation 3.1 below:

$$\text{Moisture uptake (\%)} = \frac{\text{Wa}-\text{Wb}}{\text{Wb}} \times 100\% \quad (3.1)$$

Where W_a and W_b are the weight of the specimens after and before moisture exposure. The thickness of swelling percentage for specimen calculated using equation 3.2 below:

$$\text{Thickness swelling (\%)} = \frac{\text{Ta}-\text{Tb}}{\text{Tb}} \times 100\% \quad (3.2)$$

Where T_b is the initial thickness, and T_a is the thickness of swelling of the specimen after exposure, the reported thickness swelling values are an average of five specimens. To describe the water absorption of material following Fickian behaviour. The diffusion coefficient of the composites was calculated using the equation below:

$$\frac{Wt}{W\infty} = 4\sqrt{\frac{Dt}{\pi h2}} \quad (3.3) \qquad D = \pi\left(\frac{kh}{4W\infty}\right) \quad (3.4)$$

At the initial absorption stage, water absorption (Wt) at time t increases linearly with $\sqrt{t}$, and $W\infty$ denotes the quantity after infinite time or maximum weight gain when the material approaches saturation point. Where the diffusion coefficient (D), the thickness of the specimens h, $W\infty$ is the maximum weight gain, and k is the slope of the initial plot $W(t)$ versus $\sqrt{t}$. After moisture absorption analysis, the specimen is dried at 80°C until it reaches constant weight to remove excess moisture in the specimens. Then all other characterisations were performed.

3.3.3 Flexural testing

Flexural tests were performed in a universal testing machine according to ASTM D790-92 standards. Universal Testing Machine model Shimadzu as shown in Figure 3.8, was used. The specimens were dried in the electric drying oven for 80°C until they reached constant weight to remove excess moisture inside the specimens before the test. For the flexural test, a three-point flexural test was followed based on ASTM D790-92 at a crosshead speed of 2mm/min, using a span of 70mm. Usually, after 30 days, conditioned WPC specimens became more fragile and ruptured during handling. Therefore, the experiment was conducted for up to 30 days to get better results. All tests were performed at room temperature with an average of five specimens.

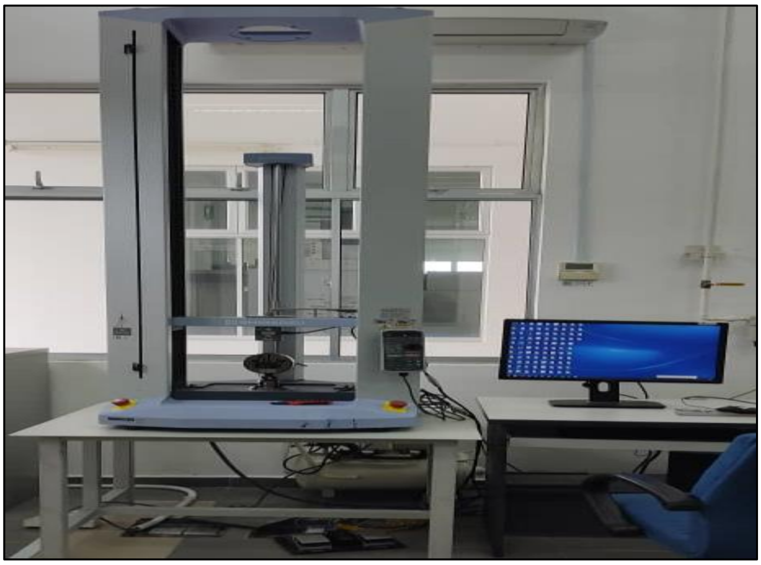


Figure 3. 8 Universal Testing Machine model Shimadzu SSM-DPN-20KN

The flexural strength calculates using equation 3.5 below:

$$\text{Flexural Strength} = \frac{3\text{LF}}{2\text{bh}^2} \quad (3.5)$$

The maximum force is F, specimen length is L, specimen width is b and specimen thickness is h. Then, the flexural modulus calculates using equation 3.6 below:

$$\text{Flexural modulus} = \frac{\text{ml}^3}{4\text{bh}^3\text{d}} \quad (3.6)$$

=

Where m is the slope, length between support from the centre l, specimen width b, specimen thickness h and deflection d. For strain at break calculated using equation 3.7

$$\text{Strain at break } (\%) = \frac{\text{La}-\text{Lb}}{\text{Lb}} \times 100\% \quad (3.7)$$

Where L_a is the specimen length increase, and L_b is the initial specimen length.

3.3.4 Morphological Analysis

The material’s interaction and morphology in terms of filler dispersion in the polymeric matrix are being assessed by imaging with the Scanning Electron Microscope (SEM) model ZEISS EVO 50 (Figure 3.9). Before taking SEM micrographs, the specimen is dried in a drying oven at 80°C until it reaches constant weight to remove excess moisture in the specimens before the test. Sputtering platinum particles into the specimen surface avoids electrical charging during the examination. The specimens are evaluated at magnification levels of 150×, 1500×, and 5000×. A Scanning Electron Microscope (SEM) was utilised to examine the fracture surface morphology of the specimens. The surface morphology of the specimen was examined at an accelerating voltage of 20kV.

Figure 3. 9 Scanning Electron Microscope model ZEISS EVO 50

CHAPTER 4

RESULT AND DISCUSSION

In this chapter, after the experiment test has been done. The result shows altogether with the discussion regarding the “Hygrothermal ageing of recycled polypropylene reinforced rubberwood flour composites for construction materials” to achieve the objective of this project.

4.1 Effect of hygrothermal ageing conditions on the moisture absorption behaviour of rubberwood flour reinforced recycled polypropylene composites

The percentage of the water absorption of the rubberwood flour reinforced recycled polypropylene in the different ageing environments is shown in Figure 4.1. It can be seen that the moisture absorption percentages in the first 14 days showed an initial straight increase followed by stabilisation after reaching equilibrium for both samples in the water immersion test and high relative humidity test, both at 50˚C. The percentages of water uptake for the WPCs depend on different periods of immersion (Babaei et al., 2014). In the water immersion test (at 50˚C), the maximum percentage of moisture uptake gain in the samples was 7.65% on day 29. At the same time, the maximum percentage of moisture uptake for the humidity test (at 90%RH with 50˚C) was 2.39% on day 30. In natural fibre polymer composites, moisture transport can be facilitated by three mechanisms: diffusion in the matrix, imperfections in the matrix and matrix interfaces (Beg and Pickering, 2008). Most water or moisture is diffuse at the interface through the capillary mechanism. In water immersion ageing, the sample is immersed entirely in the water compared to vapour exposure in 90% RH. Thus, the amount of water exposure is higher compared to vapour ageing, which results in higher water intake. Figure 4.2 also shows the same trend. It was noted that the moisture uptake percentage increased as the exposure time increased for both samples. The samples gained more weight and the percentages of moisture uptake in the immersed water at 50˚C than those exposed at 90%RH with 50˚C temperature.

The diffusion coefficient (D) of the sample from the water immersion test at 50˚C and humidity test at 90% relative humidity with 50˚C for 14 days also has been calculated and presented in Table 4.1. The diffusion coefficient characterises the ability of solvent molecules to move among the polymer segments. The sample in the water immersion test (at 50˚C) gives a higher diffusion coefficient, with a value of 0.9626 m^2/s and a high maximum moisture content. In comparison, the sample in the humidity test (at 90%RH with 50˚C) gives a lower diffusion coefficient, with a value of 0.9459 m^2/s and a lower maximum moisture content. The sample in the humidity test (at 90%RH with 50˚C) shows a slower water absorption rate than the water immersion test (at 50˚C) sample. The absorption rate is low since the humidity test sample only consists of 90% relative humidity in the humidity chamber. Compared to the water immersion test sample, which had the entire space full of water inside the water bath machine.

Wang et al., (2020) also report that the specimen's water absorption behaviour is related to wood powder content and temperature, both equilibrium moisture content and confirm that the diffusion rate of water molecules increases with the increase of wood powder content. Elevated temperatures can accelerate the diffusion rate of water molecules, thus shortening the time to reach the equilibrium moisture content. The degree of sorption and swelling obtained is determined by the liquid’s ability to interact with the cellulosic fibres (Joseph et al., 2002). Moisture diffusion into a polymer depends on its molecular and microstructural aspects, including polarity, the extent of crystallinity of thermoplastics, and residual hardeners or other water attractive species (Joseph et al., 2002).

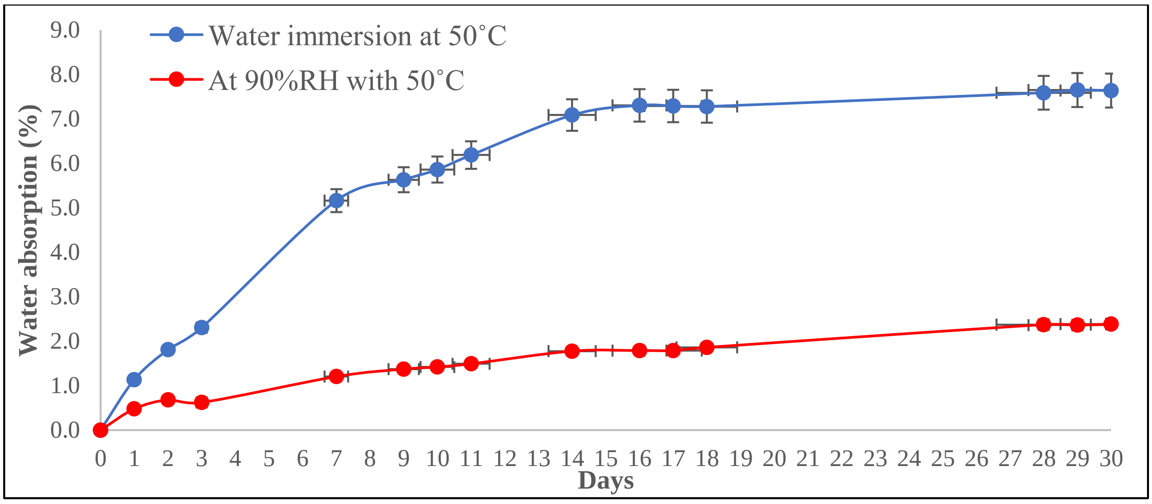


Figure 4. 1 Percentage of water absorption of the rubberwood reinforced recycled polypropylene composites for 30 days at different ageing conditions

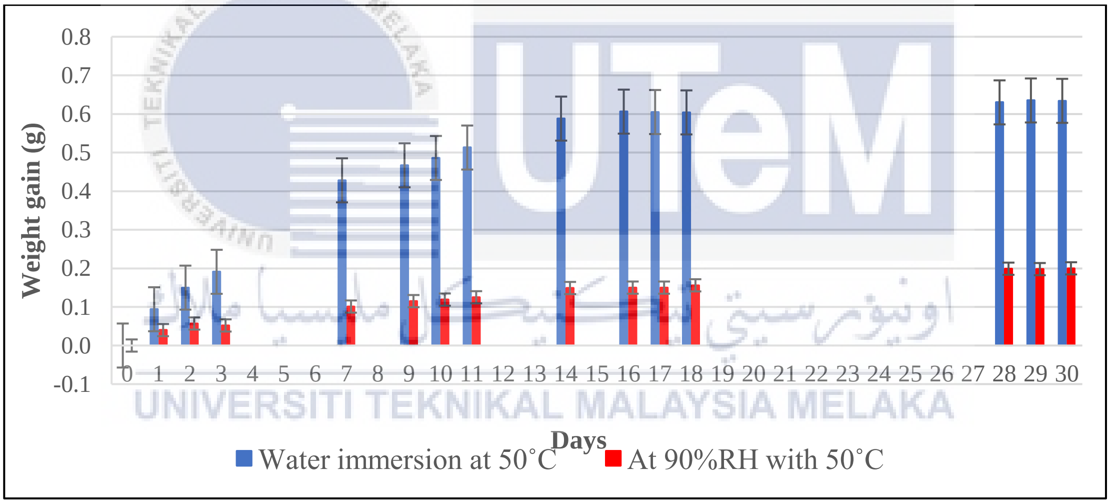


Figure 4. 2 Weight gain of the rubberwood reinforced recycled polypropylene composites for 30 days at different ageing conditions

Table 4. 1 Diffusion coefficient, D of rubberwood reinforced recycled polypropylene composites at different ageing conditions

Ageing condition	Diffusion coefficient, D
Water immersion test at 50˚C	0.9626 m^2/s
Humidity test at 90%RH with 50˚C	0.9459 m^2/s

4.2 Effect of hygrothermal ageing conditions on physical properties of rubberwood flour reinforced recycled polypropylene composites

Thickness swelling is among the most critical characteristics of composites to have a practical service life for construction material under environmental conditions. Generally, the composite's water absorption and thickness swelling increase with increased exposure time until equilibrium conditions are reached (Babaei et al., 2014). Figure 4.3 shows the percentage of changes in the thickness of the sample under the hygrothermal ageing test in different conditions. The humidity test is 90% RH at 50˚C, and the water immersion test is immersed in distilled water at 50˚C. Similar to weight gain, the thickness swelling of WPC increased with increasing exposure time.

The time to reach maximum thickness swelling in the samples was 11 days. For water immersion (50˚C), the maximum percentage of swelling thickness is 5.160% on day 30. For the humidity test (90%RH at 50˚C), the maximum percentage of swelling thickness is a 2.700% thickness increase than the original thickness on day 30. At humidity, the test shows a slowed thickness swelling trend from the water immersion test that firstly shows an aggressively increase in swelling thickness. After 12 days, both conditions showed a thickness swelling rate slowly until the end of the test 30 days. It happens due to both samples probably have reached equilibrium moisture content. By comparing, the humidity test (90% RH at 50˚C) representing water in the vapour state had significantly lower thickness swelling properties than the water immersion test (50˚C) that immersed the sample in water. Also, the sample's water absorption and thickness swelling increased due to wood flour with hydrophilic behaviour inside composites content. When exposed to moisture or water, the diffusion of water molecules into the polymeric matrix and wood flour interface occurs, resulting in changes in sample dimension. The water absorption rate that causes sample dimension changes depending upon the condition of the sample's test.

Singh et al., (2021) reported that in the study of water behaviour of jute reinforced polymer that the time to reach maximum thickness swelling in the samples was 20 days (equilibrium moisture content). The difference in time to reach maximum thickness swelling is due to the jute fibre being treated with alkali treatment, so the time to reach maximum thickness swelling in the samples is slower than in this study. They also report a similar pattern in the same pattern of moisture absorption behaviour graph as reported in this study.

When WPCs are exposed to moisture, water penetrates and adheres to the hydrophilic groups of the fibres, creating hydrogen bonds between the molecules and the fibres and reducing the adhesion of the fibre-matrix interface. The degradation process occurs when the swelling of cellulose fibres exerts pressure on the interface area leading to a micro-fracture mechanism in the matrix around the swollen fibres, which promotes capillaries and transport through micro-fractures. Water-soluble materials begin to permeate the fibres and eventually lead to final dehydration between the fibres and the matrix, as reported on flax fibres reinforced with unsaturated polyester composites (Dhakal et al., 2007). Natural fibre-reinforced composites face adverse effects when exposed to moisture. It causes a decrease in mechanical properties, provides the necessary condition for biodegradation, and changes their dimensions (Beg and Pickering, 2008; Chen et al., 2009; Wang et al., 2005, 2006). Temperature also plays a significant role in the dimensional stability of natural fibre composites. It causes direct thermal expansion or contraction by affecting the rate and moisture adsorption, leading to fibre swelling (Wang et al., 2005).

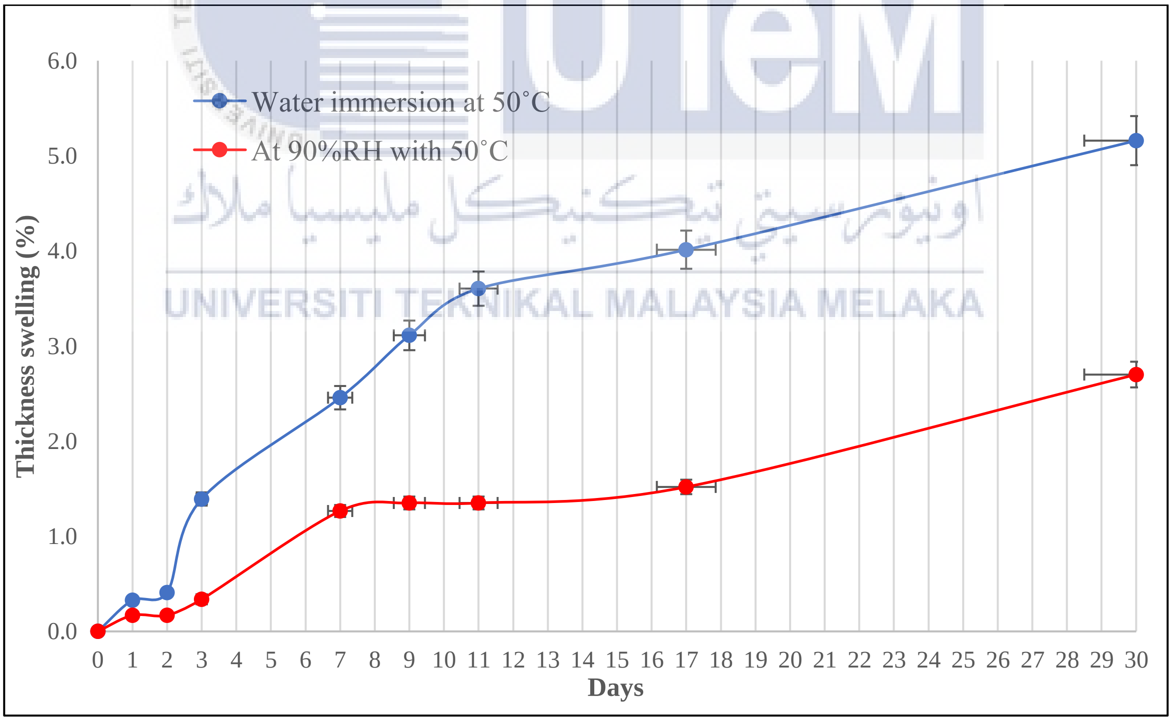
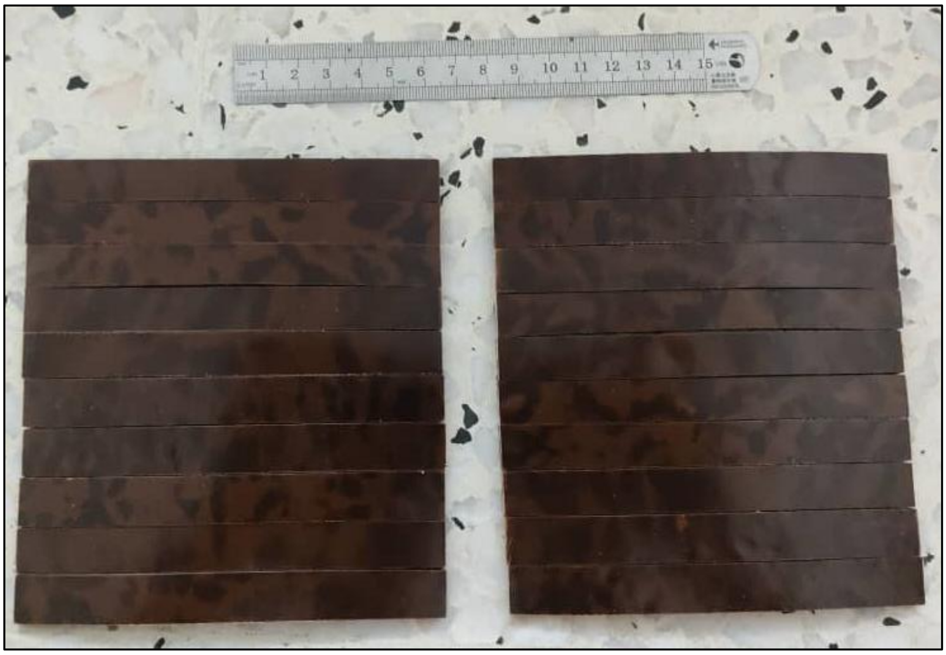


Figure 4. 3 Percentage of thickness swelling of rubberwood reinforced recycled polypropylene composites for 30 days at different ageing conditions

Figures 4.4 show the appearance of the unaged and aged samples under different environmental conditions. After undergoing hygrothermal ageing, there is a significant change in both samples in terms of colour and shape. Water immersion at 50˚C samples (Figure 4.4(b)) showed a significant change in colour after the test compared to ageing at 90% RH with 50°C samples tested in vapour (Figure 4.4 (c)) and unaged samples (Figure 4.4 (a)). Observation also showed that samples tested in vapour show a noticeable change in shape due to the thermal applied to the sample, which is 50˚C above Tg temperature for the PP has slightly straightened the sample that a bit bend before the test. Also, deposition of white chalky material was observed on the surface. It was caused by water-soluble material in the sample starting to leach during the test. Benini et al., (2011) also show the same result and stated that lignin and water-soluble products leach from the samples during the weathering process, which is apparent due to colour fading.

A significant feature of WPC is its aesthetic value which becomes compromised by weathering through severe discolouration. After hygrothermal ageing, both samples' colours become fading. Rubberwood fibre in the composites during ageing from brown to faded brown as the time of ageing increases. This discolouration results from the loss of methoxyl content of lignin. Faster fading of composites was observed with the combination of water immersion with high temperatures, which accelerates oxidation reaction. Michel and Billington (2012) agreed with the result. They stated that wood exposed to weathering initially becomes yellow and brown due to the photo-oxidation of lignin and then grey due to the degraded lignin product leaching. The study from Assarar et al., (2011) also showed that for both natural and accelerated weathering, longer exposure time increased the degree of colour change and lightness, carbonyl concentrations, and wood loss on weathered WPC surfaces.



(a) Rubberwood reinforced recycle polypropylene composites before the ageing test

(b) Ageing at 90% RH with 50°C

(c) Ageing in water immersion at 50°C

Figure 4. 4 Surface appearance of the unaged and aged rubberwood reinforced recycled polypropylene composites under different ageing conditions

4.3 Effect of hygrothermal ageing conditions on the flexural properties of rubberwood flour reinforced recycled polypropylene composites

The flexural strength and flexural modulus are significant in the criteria for using WPCs in various applications, especially for construction material. In this study, WPCs' flexural properties with three alternative conditions were investigated: unaged, humidity test (at 90%RH with 50˚C), and water immersion test (at 50˚C). Experimental results were obtained from five replicates for each case during flexural testing. Figure 4.5 shows the typical stress-strain diagram for the composites upon exposure to affect the different ageing environments.

It can be observed that the initial strength of the unaged composites is 30.49 MPa (Figure 4.6(a)). After exposure to vapour at 90% RH and 50 °C, their initial is reduced to 27.85 MPa and further to 26.61 MPa for the water immersion test. Indicated that the strength of the composites is significantly altered when exposed to moisture. The stiffness of the composites is also shown a similar pattern. Figure 4.6 (b) shows the unaged sample has the highest flexural modulus with a value of 17.01 MPa, followed by the humidity test sample with a value of 14.41 MPa and the lowest water immersion sample with a value of 11.97 MPa.

The bond's quality largely dictates a composite's strength at the interface, allowing stress transfer from the matrix to the filler. Improving the bonding at the interface results in an improvement in strength. The water absorption by the composite's wood component may cause the wood to swell. It could deteriorate the interfacial quality, resulting in decreased composites strength. Stark and Gardner (2008) also confirm that the water-soak portion of the cycling process had the most significant impact on flexural properties. After exposure to the hygrothermal ageing means after applying moisture, water and temperature to the rubberwood reinforced recycled polypropylene, the flexural properties of the composites decreased. Stark (2001) also confirmed that an increase in wood moisture generally causes a decrease in modulus. When the modulus of one component of the composites decreases, the modulus of the whole composites decreases. The high moisture content reduces the mechanical properties of the composite, especially the flexural properties.

In contrast to their flexural strength and modulus, the strain at break for the composites increases after moisture exposure. Figure 4.6 (c) shows that the initial strain at break for rubberwood reinforced recycled polypropylene composite is 1.83 %. After exposure to high humidity (90% RH) at 50 °C, the strain at break value is increased to 3.34% and a further 3.94% for samples in water immersion. Suggests that the composites become more ductile after exposure to moisture. Cai et al., (2022) suggested that this increase in the strain at break is due to plasticising effect of the amorphous region of the natural fibre and re-orientation of the cellulose microfibrils due to water-induced. The result of the flexural properties of the rubberwood reinforced recycled polypropylene is summarised in Table 4.2.

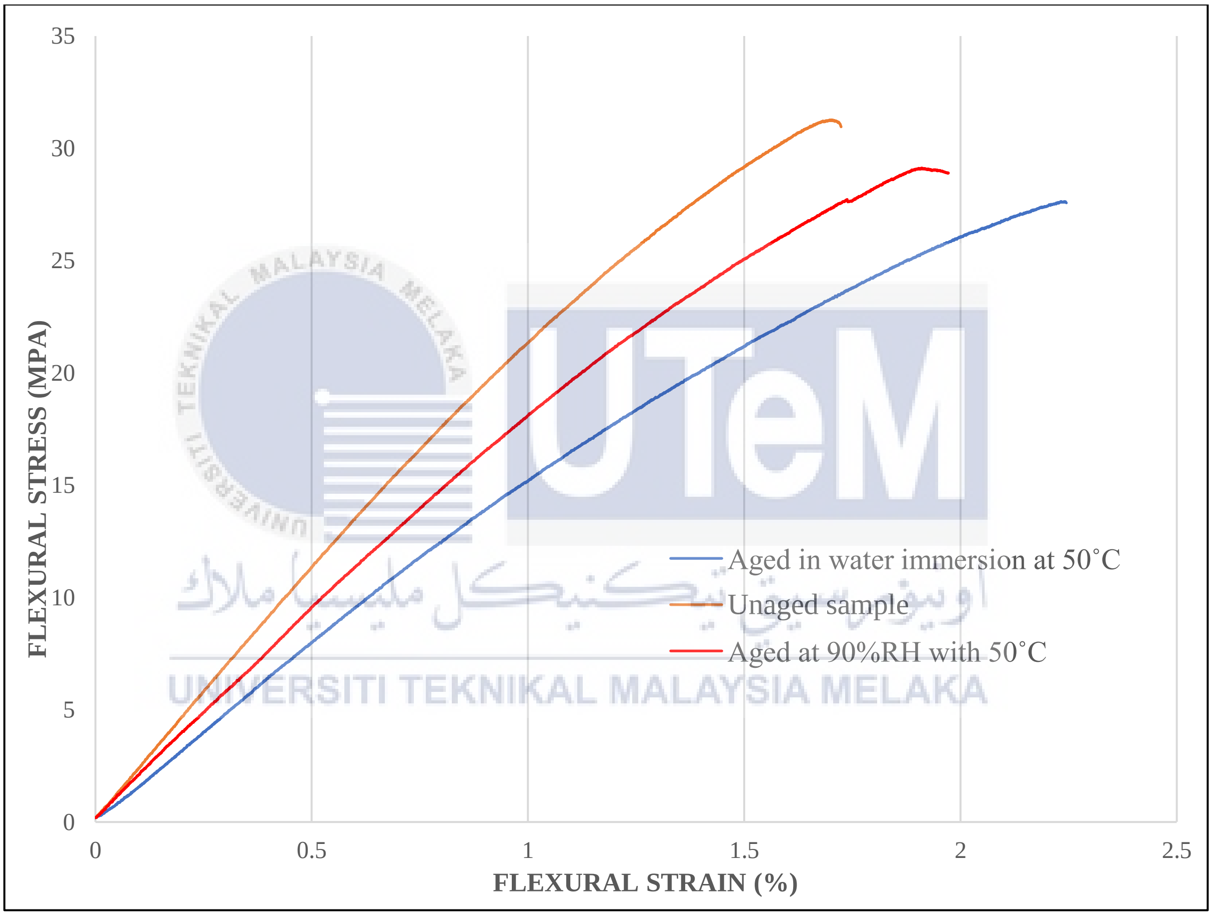
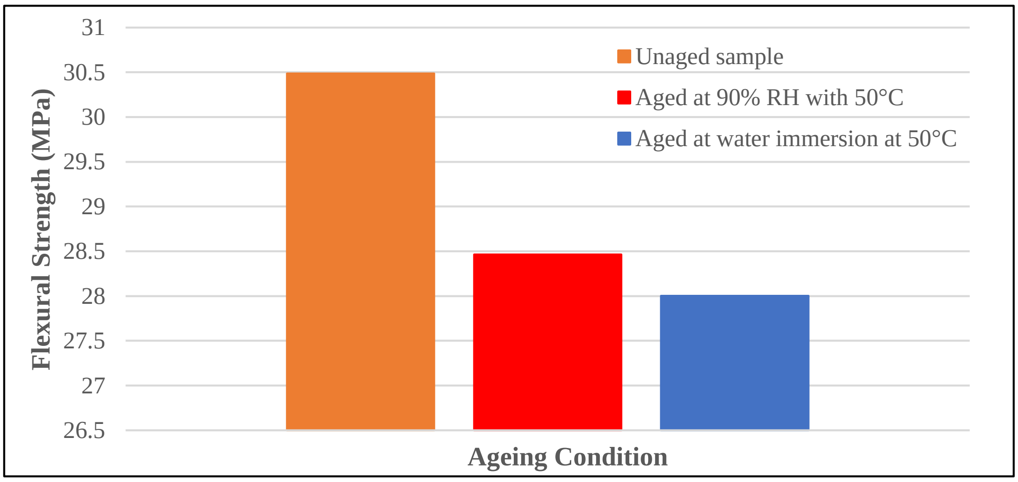
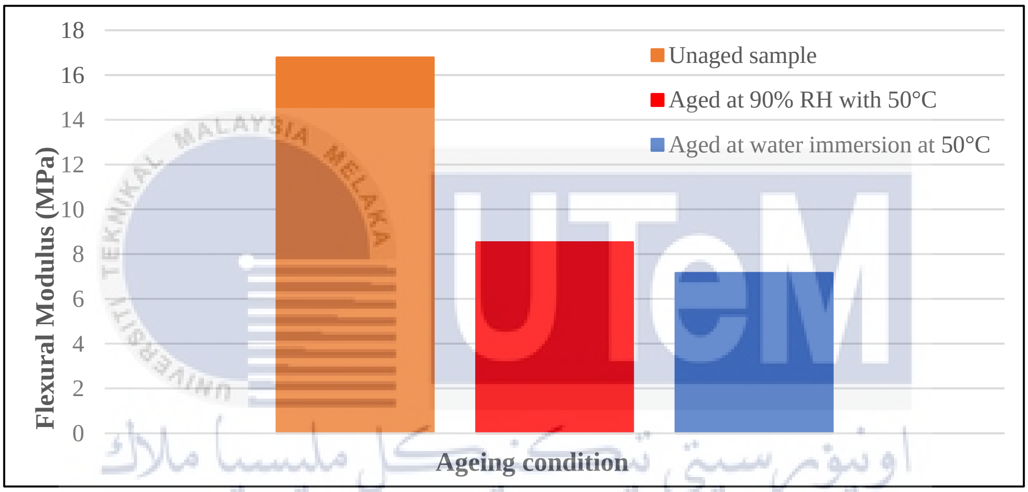


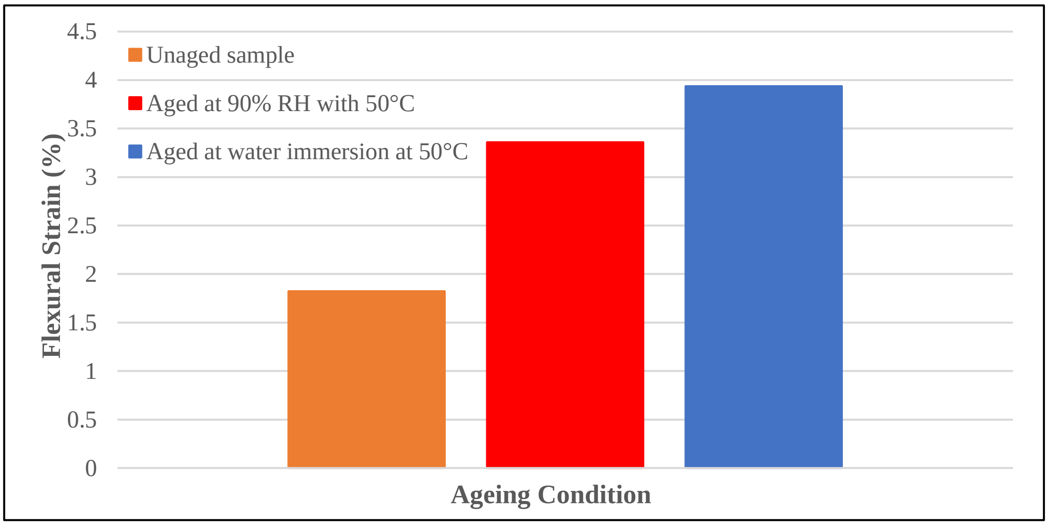
Figure 4. 5 Typical stress-strain diagrams for unaged and aged rubberwood reinforced recycled polypropylene composites



(a) Flexural strength of the unaged and aged rubberwood/rPP composites



(b) Flexural modulus of the unaged and aged rubberwood/rPP composites



(c) Flexural strain of the unaged and aged rubberwood/rPP composites

Figure 4. 6 Flexural properties of the unaged and aged rubberwood reinforced recycled polypropylene composites under different ageing conditions

Table 4. 2 Flexural properties of unaged and aged rubberwood reinforced recycled polypropylene composites

Sample condition	Unaged sample	Aged at 90% RH with 50°C	Aged at water immersion at 50°C
Flexural Strength (MPa)	30.486±0.53%	28.464±0.82%	28.003±1.41%
Flexural Modulus (MPa)	16.786±1.10%	8.526±0.67%	7.162±0.84%
Flexural strain at break (%)	1.822±0.11%	3.359±0.34%	3.939±0.34%

4.4 Effect of hygrothermal ageing conditions on the surface fractured of rubberwood flour reinforced recycled polypropylene composites

Figure 4.7 shows the overview of the fracture surface of the rubberwood reinforced recycled polypropylene composites after the flexural test. From left to right is an unaged sample, aged at 90% RH with 50˚C sample and aged in water immersion at 50˚C sample. This surface fracture has been examined using a scanning electron microscope (SEM).

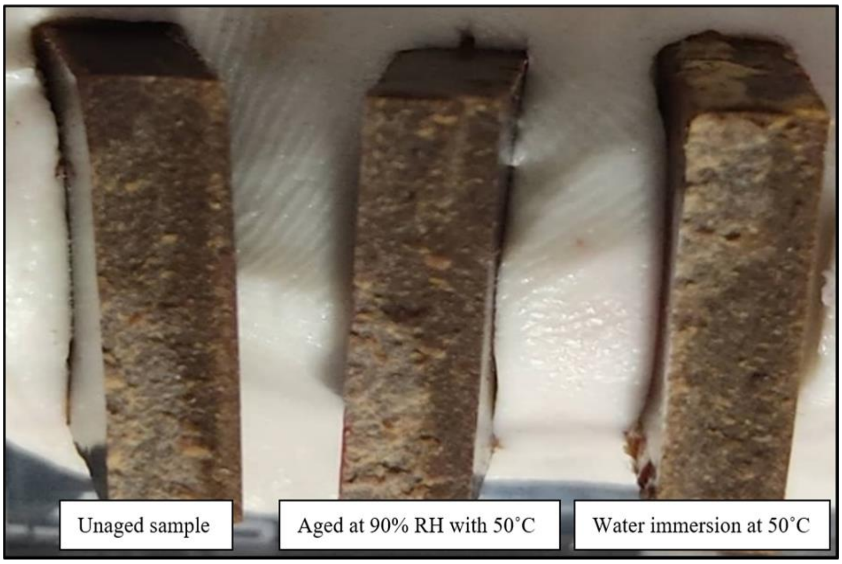
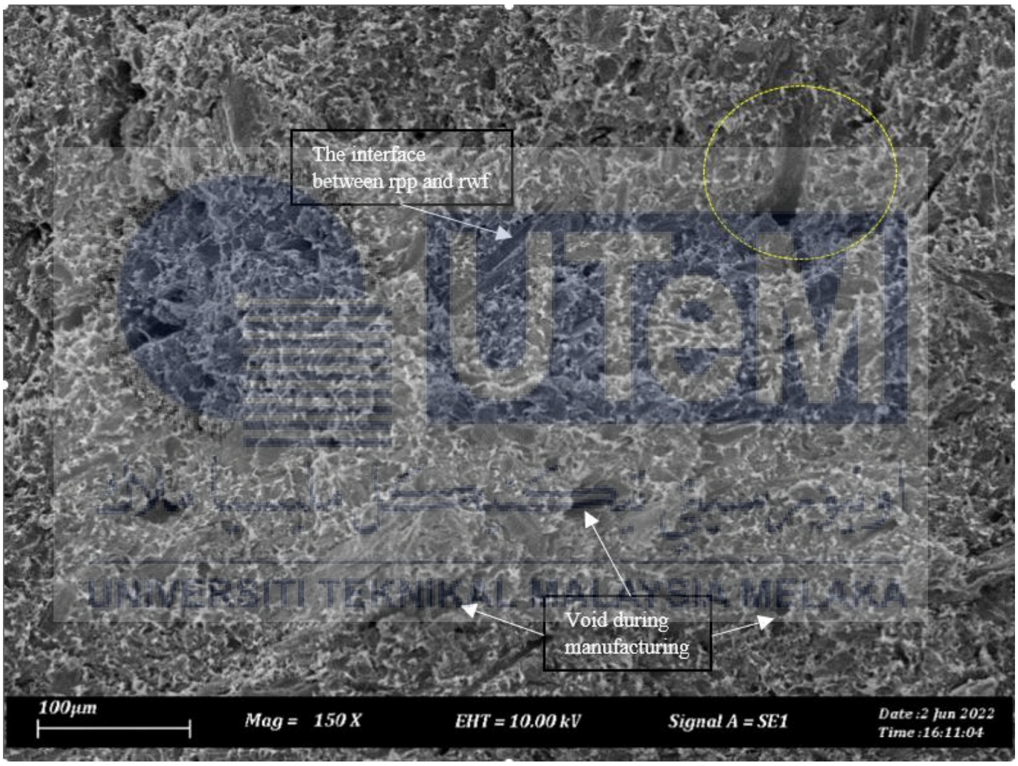
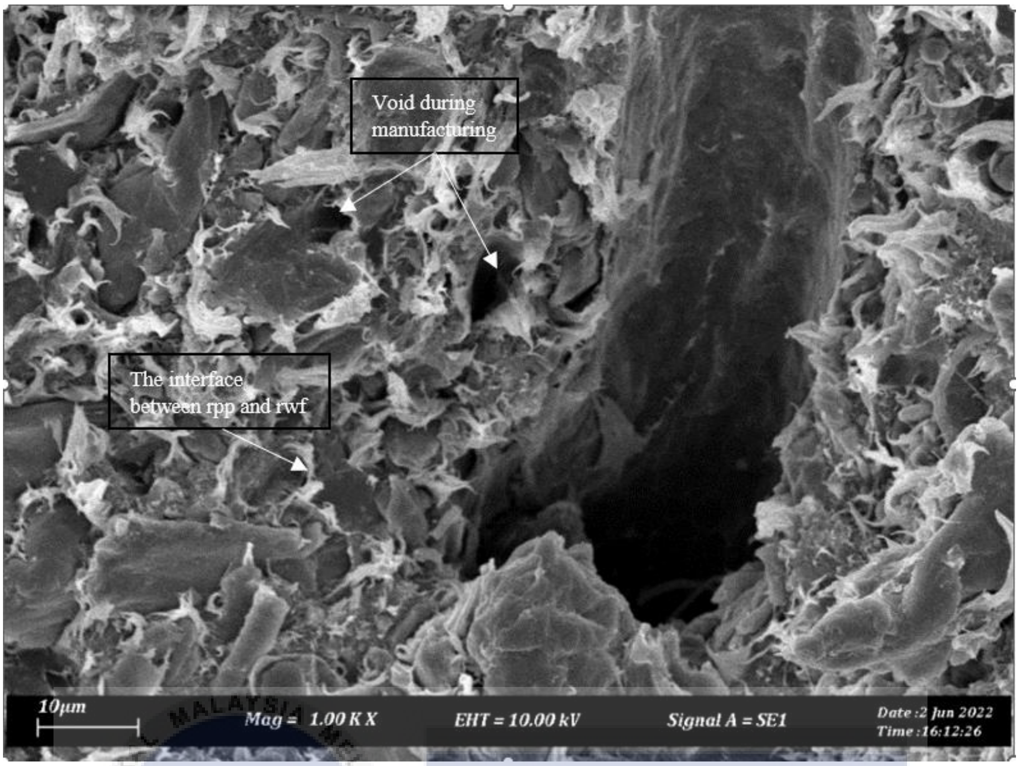


Figure 4. 7 Flexural fractured surfaces of rubberwood reinforced recycled polypropylene composites at different ageing conditions

Figures 4.8 till 4.10 show the composites morphology analysis after exposure to different ageing conditions. The fractured surface of the unaged sample in Figures 4.8 (a) and (b) shows that interfaces between rubberwood fibres and the recycled polypropylene matrix have been conserved after the flexural test. This means that no debonding and no pull-out of fibres are observed. Moreover, the presence of voids caused by the manufacturing process is detected in Figures 4.8 (a) and (b). Based on the SEM image result, fracture of the unaged sample is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the fracture of rubberwood fibre that causes a brittle fracture.



(a) Unaged rubberwood reinforced recycled polypropylene composites at 150×mag



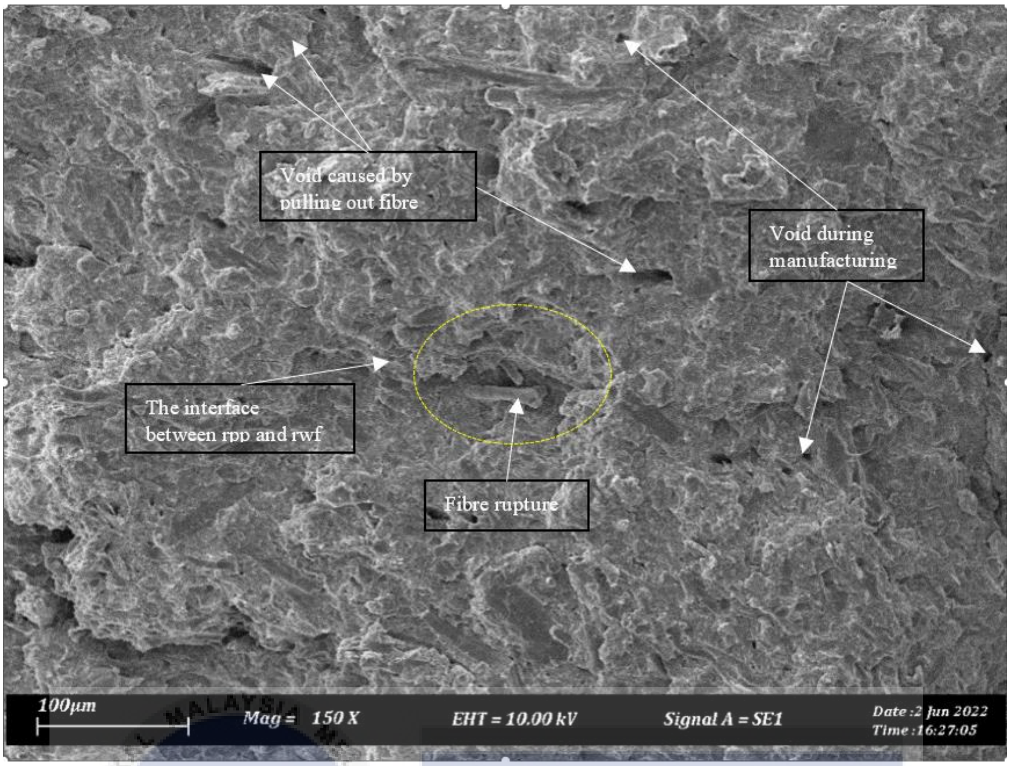
(b) Unaged rubberwood reinforced recycled polypropylene composites at 1000×mag

Figure 4. 8 Flexural fractured surface of the unaged rubberwood reinforced recycled polypropylene composites

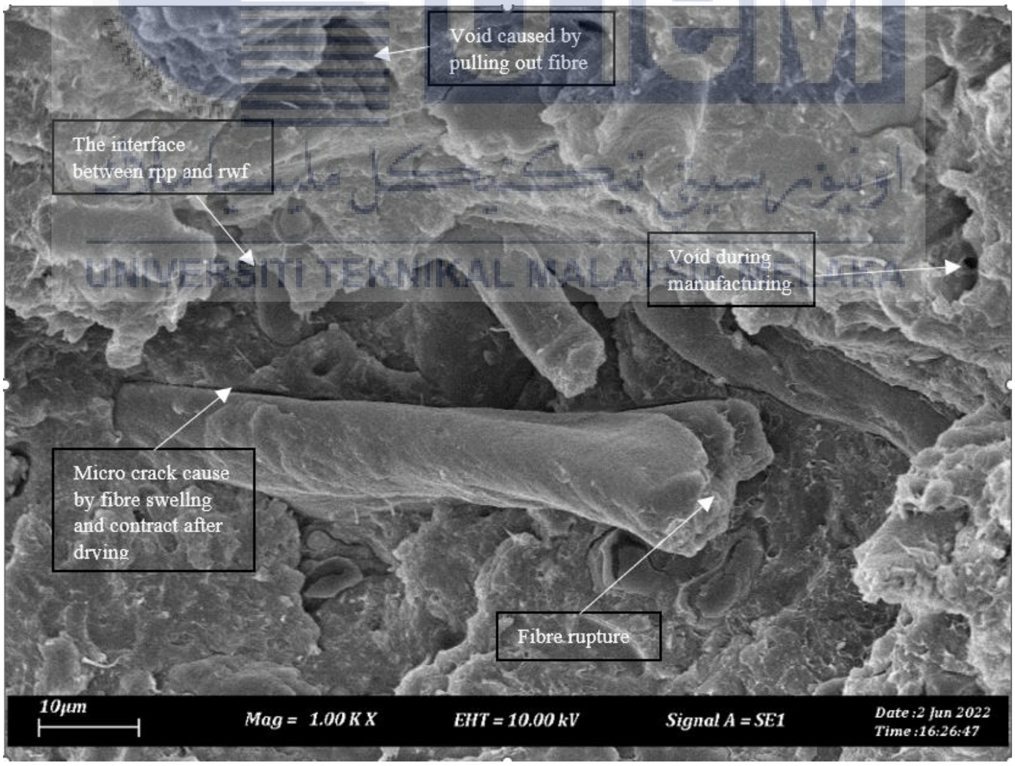
Figure 4.9 shows the fracture surface of the rubberwood reinforced recycled polypropylene composites after exposure for 30 days, aged at 90% RH with 50˚C. It can be seen that the composites fracture in a ductile manner. As shown in Figures 4.9 (a) and (b), performing a flexural test after hygrothermal ageing induces fibres debonding and voids caused by the pull-out of fibres. Based on a comparison between Figure 4.8 and 4.9, debonding and pull-out of some fibres in the aged sample are caused by the reduction of interfacial adhesion between the fibres and the matrix. The strength of the fibre reduces and causes the fibre to rupture during the flexural test. Fracture of the humidity test sample is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the swelling and shrinking rubberwood fibre that causes a ductile fracture. In this case, representing water in the vapour state causes the swelling of fibres during water uptake to initiate a micro crack in the sample and by the pressure of water inside voids in the matrix caused by the manufacturing process.

Figure 4.10 shows the fracture surface of the rubberwood reinforced recycled polypropylene composites after exposure to 30 days of water immersion at 50˚C. It can be seen that the fracture of the composite is in a high ductility manner. As shown in figures 4.10 (a) and (b), performing a flexural test after hygrothermal ageing induces fibres debonding and voids caused by the pull-out of fibres. Comparison between Figures 4.8, 4.9 and 4.10, debonding and pulling out of fibres in the aged sample are caused by the reduction of interfacial adhesion between the fibres and the matrix. After immersion in water at 50˚C for 30 days, severe splitting of the fibres was observed in Figure 4.10 (b). Fracture of the water immersion test sample is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the fracture of RWF fibre that causes a high ductile fracture. In this case, water causing the swelling of fibres during water uptake initiates a micro crack in the sample. Drying after the ageing, the swelled fibre shrinks, leading to a bigger micro-crack.

In summary, the unaged sample fracture surface shows that interfaces between fibres and the matrix have been conserved after the flexural test. No debonding and no pull-out of fibres are observed. However, the presence of voids caused by the manufacturing process is detected. Fracture is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the fracture of RWF fibre that causes a brittle fracture. The fracture was ductile for the sample aged at 90% RH with 50˚C. The fibre rupture was observed. The water vapour state causes the swelling of fibres during water uptake to initiate a micro crack in the sample and by the pressure of water inside voids in the matrix caused by the manufacturing process. For the sample aged in water immersion at 50˚C, the fracture of the composite is highly ductile. Severe splitting of the fibres was observed. Water causing the swelling of fibres initiates a micro crack in the sample, then drying after the test shrinks the swelled fibre leading to a bigger micro-crack. For the aged sample, performing a flexural test after hygrothermal ageing induces fibres debonding and voids caused by the pull-out of fibres. Debonding and pulling out of some fibres are caused by the reduction of interfacial adhesion between fibres and matrix. Joseph et al., (2002.) studied also agree that with the absorption of water, hydrogen bonds between water molecules and the cellulose fibres are formed, causing loss of compatibilisation between the cellulose fibre and the matrix which results in debonding and weakening of the interface adhesion. Water causing the swelling of fibres during water uptake initiates a micro crack in the sample and then drying after the test shrinks the swelled fibre leading to crack and fracture is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the fracture of rubberwood fibre.

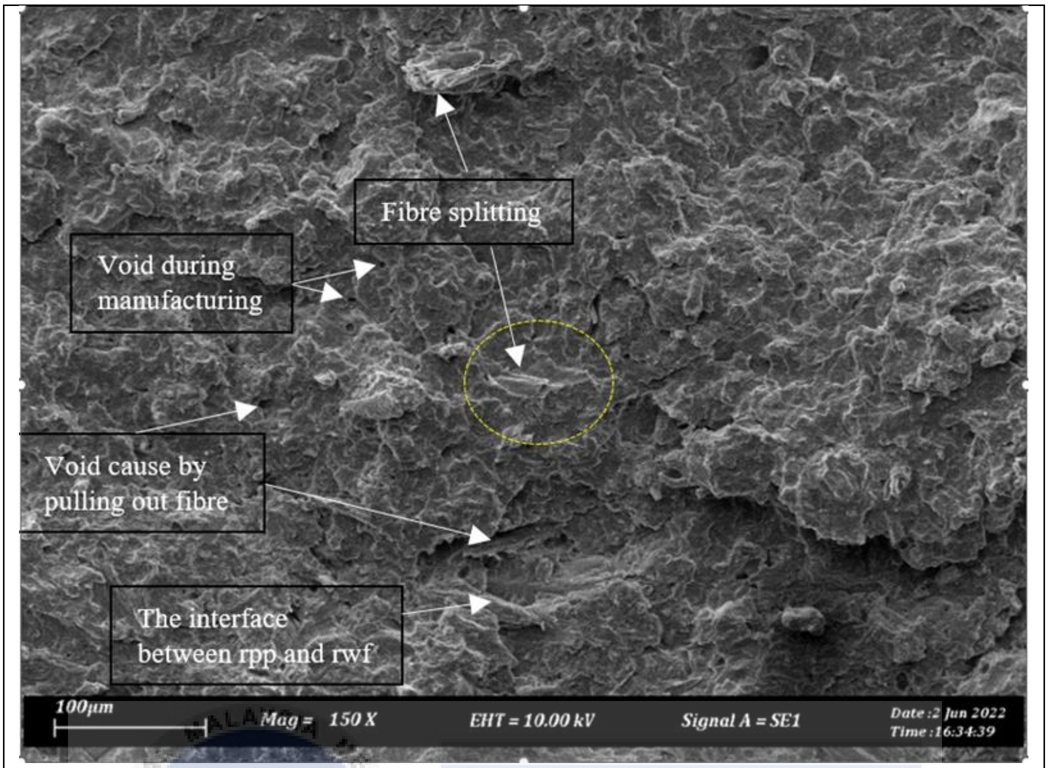


(a) Aged at 90%RH with 50˚C at 150×mag

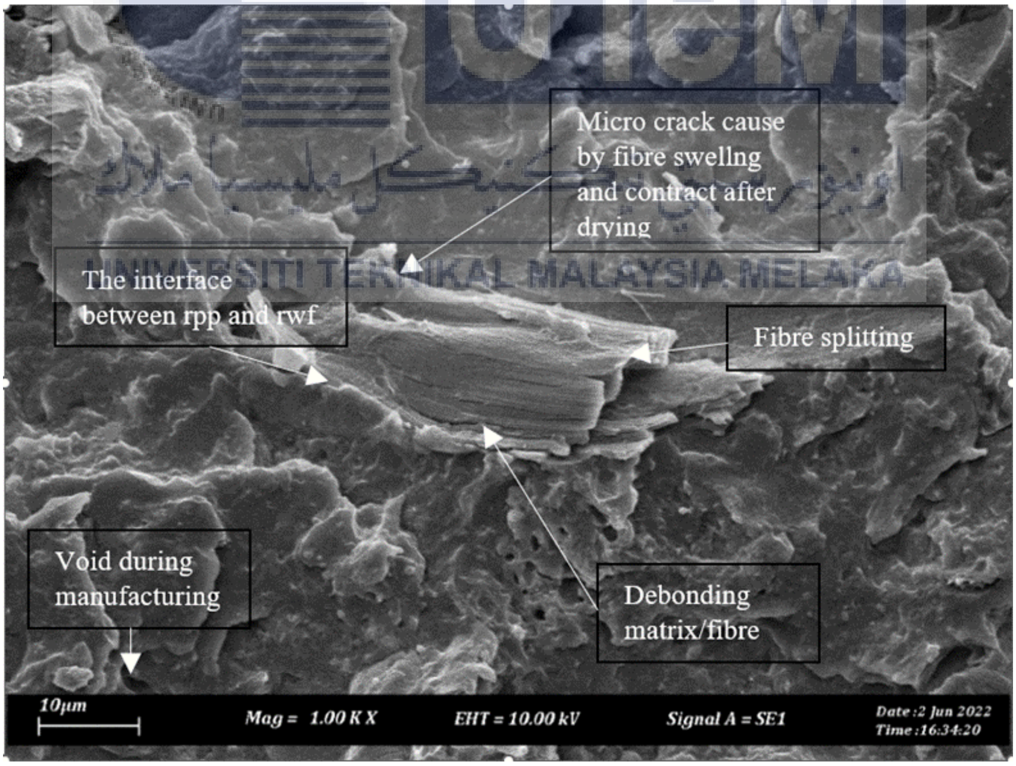


(b) Aged at 90%RH with 50˚C at 1000×mag

Figure 4. 9 Flexural fractured surface of rubberwood reinforced recycled polypropylene composites aged at 90%RH with 50˚C



(a) Aged in water immersion with 50˚C at 150×mag



(b) Aged in water immersion with 50˚C at 1000×mag

Figure 4. 10 Flexural fractured surface of the rubberwood reinforced recycled polypropylene composites aged at water immersion at 50˚C

CHAPTER 5

CONCLUSION AND RECOMMENDATION

In this chapter, the conclusion of the results of the hygrothermal ageing of rubberwood flour reinforced recycled polypropylene composites for construction material is revealed. This chapter includes recommendations and suggestions for further research.

5.1 Conclusion

The first objective of this study is to compare the effect of hygrothermal ageing on the physical properties of rubberwood reinforced recycled polypropylene composites. The conclusions follow:

a) The water absorption behaviour of the rubberwood reinforced rPP composites upon exposure to moisture for 30 days following the Fickian behaviour.

b) The water absorption rate in water immersion is higher than vapour exposure at 90% RH at 50°C, with their diffusion coefficient of 0.9626 m^2/s and 0.9459 m^2/s, respectively.

c) The thickness swelling after exposure to water immersion and vapour exposure at 90% RH at 50°C is 5.16% and 2.70%, respectively. Indicating the composites have poor dimension stability in a moisture-rich environment.

The second objective of this study is to compare the effect of hygrothermal ageing on the flexural properties of rubberwood reinforced recycled polypropylene composites. The conclusions follow:

a) The initial strength of the rubberwood reinforced rPP is 30.49 MPa. Upon exposure to humidity at 90% RH and elevated temperature (50˚C), the flexural strength of the composites is reduced to 27.85 MPa and further reduced to 26.61 MPa upon exposure to 30 days of water immersion at 50°C.

b) The initial modulus of the rubberwood reinforced rPP is 17.01 MPa. Upon exposure to humidity at 90% RH and elevated temperature (50˚C), the flexural modulus of the composites is reduced to 14.41 MPa and further reduced to 11.97 MPa upon exposure to 30 days of water immersion at 50°C, indicating the deleterious effect of moisture onto the flexural strength and modulus of the composites.

c) In contrast, the strain at the break of the composites increases upon exposure to moisture. The initial strain at the break of the composites is 1.83%. It increases to 3.34% and 3.94% when exposed to moisture in vapour exposure at 90% and water immersion, respectively. Suggesting the composites becomes ductile due to plasticising effect of the polymer and amorphous region of the fibres.

The third objective of this study is to evaluate the fracture surface of rubberwood reinforced recycled polypropylene composites caused by hygrothermal ageing. The conclusion is as follows:

a) The composites failed due to fibre and matrix fractured in the unaged sample. No significant fibre pull-out is observed. But the presence of voids caused by the manufacturing process was detected. It concludes that fracture of the unaged sample is caused by the initiation and propagation of micro-cracks in the rPP matrix and by the fracture of rubberwood fibre that causes a brittle fracture.

b) Upon exposure to the vapour in 90%RH with 50˚C, the surface of the composites showed the present fibres debonding and voids. Ageing causes the weakening of fibre and matrix interface. It is indicated by the presence of the pull-out of fibre which supported the finding of the flexural properties.

c) In the water immersion aged composites, similar fracture behaviour is observed but with the significant presence of the pull-out fibre with the addition of microcracking and a gap between the fibre and matrix. It indicates the weakening of the fibre and matrix interface, which results in lower strength and modulus value.

5.2 Recommendation

Based on the study, much improvement can be made for future development in studying rubberwood flour reinforced recycled polypropylene composites for construction material. The recommendation for improvement is as follows:

a) The whole surface of rubberwood flour reinforced recycled polypropylene composites should be fully applied with coating before the ageing and test the effect of the type of coating on the properties of the composites. If the surface of the composites is coated fully during the ageing, the moisture absorption may be low since the surface of the composites is exposed after the cut or manufactured. It sealed the composites from the moisture and heat and mimicked their actual behaviour in the application.

b) Fatigue testing can be carried out to determine the behaviour of the composites under cyclic conditions.

c) Wood polymer composites also may be susceptible to changes due to load and thermal combination; thus, is it suggested to carry out the creep test.

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