

MECHANICAL PROPERTIES OF POLYPROPYLENE MIXED WITH
RICE HUSK POLYMER COMPOSITES UPON ULTRAVIOLET
IRRADIATION

NUR AFIQAH BINTI SUFIAN

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Faculty of Mechanical Engineering and Manufacturing Universiti Tun Hussein Onn
Malaysia

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ABSTRACT

Wood Polymer Composites (WPCs), a material made from polymer matrix and natural fiber are growing into a high market industry due to it being sustainable and more environmental friendly which acts as an alternative to polymer. This study aims to investigate the influence of weight percentage of rice husk (RH), recycled polypropylene (WPP) and homopolymer polypropylene (HPP) mixture affects the mechanical characteristics and the photodegradation of this material particularly after being subjected to UV irradiation. In this study, WPC specimens were prepared by mixing the WPC pellets (WPP mixed with RH) obtained from the industry with 10%, 20% and 30% of (HPP) respectively through injection molding. The percentage of WPC pellets were 100% WPP, (80% WPP + 20% RH), (60% WPP + 40% RH) and (40% WPP + 60% RH). The specimens were later exposed to 1000, 2000, 3000, 4000, 5000 and 6000 hours of UV irradiation under accelerated weathering. The specimens were then subjected to tensile test, flexural test, FTIR analysis and morphological studies of the fractured surface by means of an optical microscope. In this thesis, WPP, RH, and HPP were denoted by the letter A, B, and C, respectively. The results obtained show that specimen A₄₀B₆₀C₀ has the highest modulus of elasticity (MOE) and carbonyl index (CI) value but lowest flexural strength. Specimen A₆₀B₄₀C₃₀ however exhibits the highest value for flexural modulus and the lowest value for carbonyl index. It can be concluded that WPC specimens exhibit decrease in mechanical properties after they were exposed to UV irradiation. CI values obtained from FTIR analysis confirmed this. The morphological studies of the fractured surfaces are also in agreement whereby formation of voids was detected on specimens after UV irradiation. The increased content of rice husk contributed to the decrease of mechanical properties. This effect was counter with the addition of HPP into the composition by reducing the exposure of rice husk fillers to UV irradiation and humidity and increasing the interface bonding of rice husk and polymer matrix.

ABSTRAK

Komposit Polimer Kayu (WPC), adalah bahan yang diperbuat dari matriks polimer dan serat semula jadi. Ianya sedang berkembang pesat menjadi industri pasaran yang tinggi kerana lebih lestari dan mesra alam serta bertindak sebagai alternatif kepada polimer. Kajian ini adalah bertujuan untuk mengkaji pengaruh peratusan berat sekam padi (RH), campuran polipropilena kitar semula (WPP) dan homopolimer polipropilena (HPP) ke atas ciri mekanikal dan fotodegradasi bahan ini terutamanya setelah terdedah kepada sinaran ultraviolet. Dalam kajian ini, spesimen WPC disediakan dengan mencampurkan pelet WPC (WPP dicampur dengan RH) yang diperoleh dari industri dengan 10 %, 20 % dan 30 % wt% dari (HPP) masing-masing menggunakan pengacuan suntikan. Peratusan pelet WPC adalah 100 wt% WPP, (80 wt% WPP + 20 wt% RH), (60 wt% WPP + 40 wt% RH) dan (40 wt% WPP + 60 wt% RH). Spesimen-spesimen tersebut kemudian didedahkan kepada 1000, 2000, 3000, 4000, 5000 dan 6000 jam sinaran UV di bawah cuaca yang dipercepatkan. Spesimen tersebut kemudian menjalani ujian tegangan, ujian flexural, analisis FTIR dan kajian morfologi permukaan yang patah menggunakan mikroskop optik. Dalam tesis ini, WPP, RH, dan HPP masing-masing diwakili oleh huruf A, B, dan C. Hasil yang diperoleh menunjukkan bahawa spesimen $A_{40}B_{60}C_0$ mempunyai nilai modulus keanjalan (KPM) dan indeks karbonil (CI) tertinggi tetapi kekuatan lenturan terendah. Spesimen $A_{60}B_{40}C_{30}$ bagaimanapun menunjukkan nilai tertinggi untuk modulus lenturan dan nilai terendah untuk indeks karbonil. Spesimen WPC menunjukkan penurunan sifat mekanikal setelah mereka terdedah kepada sinaran ultraviolet. Peningkatan kandungan sekam padi menyumbang kepada penurunan sifat mekanikal. Kesan ini dapat dibendung dengan penambahan HPP ke dalam komposisi dengan mengurangkan pendedahan sekam padi kepada sinaran ultraviolet dan kelembapan serta meningkatkan ikatan antara permukaan sekam padi dan polimer.

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LIST OF SYMBOLS AND ABBREVIATION

WPCs	-	Wood Polymer Composites
PE	-	Polyethylene
PET	-	Terephtalate
HDPE	-	High-Density Polyethylene
PVC	-	Polyvinyl Chloride
LDPE	-	Low-Density Polyethylene
PP	-	Polypropylene
PS	-	Polystyrene
RH	-	Rice Husk
MAPP	-	Maleic Anhydride
IBS	-	Industrialized Building System
UV	-	Ultra-Violet
FTIR	-	Fourier Transform Infrared
OM	-	Optical Microscopy
CI	-	Carbonyl Index
VE	-	Vinyl Esters
IM	-	Injection Moulding
MPa	-	Mega Pascal
HIPS	-	High Impact Polystyrene
ABS	-	Acrylonitrile Butadiene Styrene
PA	-	Polyamide
PC	-	Polycarbonate
HPP	-	Homopolymer Polypropylene
RCP	-	Random Copolymer
ICP	-	Impact Copolymer
iPP	-	Isotactic Polypropylene

RPP	-	Recycled Polypropylene
RPS	-	Recycled Polystyrene
MOR	-	Modulus of Rupture
MOE	-	Modulus of Elasticity
T _g	-	Glass Transition
mm	-	millimetre
°C	-	Degree Celsius
wt%	-	Weight percentage



CHAPTER 1

INTRODUCTION

1.1 Introduction

For the past few decades, the developments of Wood Polymer Composites (WPCs), especially in terms of their application and market share, have been increasing. Many researches and studies are being conducted on this material type due to its cost efficiency and the role it plays in decreasing the environmental impacts. Generally, WPCs are composites that are comprised of polymer matrix and natural fiber. The polymer matrix can be either virgin or waste plastics. Virgin plastic is a newly created plastic resin without any recycled materials. This type of plastic is produced either by using natural gas or crude oil and is commonly used to create brand new plastic products for the very first time. The forms of virgin plastics include Polyethylene (PE), Polyethylene Terephthalate (PET), High-Density Polyethylene (HDPE), Polyvinyl Chloride (PVC), Low-Density Polyethylene (LDPE), Polypropylene (PP), and Polystyrene or Styrofoam (PS). Plastic waste refers to the accumulation of plastic objects, such as plastic bottles, in the earth's environment that is causing adverse effects on the habitats and humans in this world. Meanwhile, the natural fiber used in WPC composite as reinforcing materials is usually derived from renewable and carbon dioxide neutral resources, such as wood or plants. Examples of natural fibers include softwood radiate pine, straw, rice husk, sawdust, cotton, jute, and wool.

Today, WPCs are the most commonly used in the construction field. So far, WPCs have been shown to be compatible for indoor and outdoor uses, such as railings, fences, landscaping timbers, cladding and siding, park benches, molding and trim, window and door frames, and indoor furniture. In addition, rice husk and polypropylene-based composites are among the types of WPCs that have been gaining attention as alternative construction materials.

1.2 Background Research

Wood polymer composites are still considered a new material in the industry. Hence, there are several aspects that need to be addressed to utilize this material further. Numerous researches on WPCs are being performed since their advantages vary from structural to non-structural. Many developers are concentrating more on how to enhance the yields of this new material in the context of processing, physical characteristics, aesthetic values, and weathering effects. Overall, the benefits of WPCs as the substitution of other materials are expected to be better. The aim of developing WPCs as outdoor products is because of the durability of these compounds. Nevertheless, the knowledge of this product regarding its long-term efficiency is still vague.

Wood polymer composites are developed by the combination of natural fibers with plastic materials through the means of different plastic processing technologies. These materials are economical and have superior mechanical characteristics, as compared to the materials with synthetic fibers, depending on the differences in fibers densities. Furthermore, the use of wood from agricultural by-products (agro-waste) and recycled polymer could assist in addressing environmental concerns. Lignocellulosic-plastic composites reside in semi-crystalline plastics, such as polypropylene (PP) and polyvinyl chloride (PVC). Thermoplastics with melt temperatures of below 200°C are commonly used, due to the limitation of natural fiber thermal stability. Polypropylene-based composites utilization is mainly used in the automotive industry, but recently, various studies have been conducted to develop this material as building profiles.

The use of agro-waste is also being implemented vigorously in the making of wood-plastic composites. Examples of agro-wastes or by-products of agricultural

products include husk, fiber, cobs, and straw. One of the natural fibers suitable to be utilized in South East Asia is rice husk (RH) because rice is the most significant crop grown in this region. It was expected that the overall rice demand in South East Asia would grow to more than 160 million tonnes per year. Per capita rice consumption in Asia is far higher than in all other regions. Rahman et al. (2023) stated that on average, more than 77 kg of rice is consumed annually by every person in Asia. By the year 2020, the amount of rice husk in the landfills would also increase, as it serves no purpose after the paddy is being processed by rice milling. A study by Onoja et al. (2019) stated that RH consists of 35% cellulose, 25% hemicellulose, 20% lignin, and 17% ash (94% silica) and 3% wax by weight. Moreover, the plastic matrix can act as a moisture barrier for the natural fiber.

Since wood polymer composite itself is still considered a new material, numerous features should be deliberated, from where the material originated to designing composites consisting of RH and PP. The weak distribution of fibers into the composite occurred from the weak bonding between hydrophilic fibers and hydrophobic thermoplastic matrices. Poor adhesion bonding is instigated by the strong bonds between the fibers. Thus, stress and force could not be competently transferred to the fibers.

WPCs have a great number of advantages, namely economic, sustainable and have high material and mechanical characteristics. The key purpose of fillers usage in composites is to make it an economical and high-strength material. Consequences in terms of environmental impacts on the resistance of fiber have been produced, which has led to the rise of researches on the variation in the characteristics of WPCs when subjected to either artificial or outdoor weathering conditions. It is known that the addition of coupling agents can effectively control the photodegradation impacts of polymers. However, the impacts are less pronounced on wood materials. Still, the interest in the photodegradation of polymeric materials has been rising, mainly due to the rise of commercial macromolecules utilization. The impacts of accelerated photodegradation may affect the combination of rice husk into the polymer composites due to the fiber deterioration.

Based on a forecast made by HOSUNG (2019), the wood plastic composite market of Asia Pacific is expected to experience a significant increase, with a compound annual growth rate of 12.96% within the period of 2019 until 2027. The increasing demands in the mentioned region, alongside the rise of new manufacturing processes

and new recyclable raw materials, are contributing to the market's prospects of potential growth. The building and construction sector in China shows a prominent position of WPC application. The automotive industry in Japan, on the other hand, is utilizing the WPC with plastic composites comprising of wood, straw, rice, and several other materials. The approach aims to optimize the weight of cars and assist with carbon footprint reduction. As a result, the WPC market in India is expected to have a high compound annual growth rate due to the increasing application of materials in the building and construction, automotive parts, and various sectors (HOSUNG, 2019).

1.3 Problem Statement

As stated earlier, the applications of WPCs are increasing in various fields. Recently, a number of institutions and private sectors, especially the IBS industries in the European region, have introduced a new policy that requires the production of more environment-friendly products. This leads to an increase in production caused by the high demands. Polymers are known to be one of the contributing waste products in the world, as this compound has wide applications in various fields. The large usage mostly contributes to the increasing effects and impacts on environmental health. Some of these polymers are also derived from non-renewable sources, such as fossil fuels. The application of recycled polymer waste in WPCs might help in satisfying the demands of raw materials, decrease the environmental impacts and increase the cost-efficiency of the products. Previous studies showed that the properties of recycled polymers have insignificant differences from the virgin polymers, as these materials are made up of different contents, such as additives and coupling agents, as compared to the traditional products.

Due to the increasing hurdles in obtaining raw materials supplies for wood-based panels, developments and enhancements are being done in this particular field in the hope of diversifying the materials supplies. The incorporation of lignocellulosic materials, such as straw (Harun et al., 2022) and n-rice husk, have been successfully developed through many studies. The Food and Agriculture Organization (FOA) stated that the annual production of paddy in 2005 was 628 million tons, with an additional 1% in the following year. Rice husk can be found in abundance as a waste product

from the rice milling industry. This fiber possesses no commercial interests and may eventually lead to problems, such as soil pollution and an increase in waste landfills. Hence, the consideration to combine this natural fiber into polymer formulations might assist towards a more positive environmental effect and increase in cost efficiency. Rice husk is known to be naturally tough, insoluble in water, abrasive resistant, and can act as a structural arrangement of silica-cellulose. The chemical constituents of rice husks samples may differ from one another, contributed by the types of paddy, geographic conditions, climate variations, the chemistry of soil, and fertilizers used.

Weathering is another aspect to be considered, especially for the outdoor application of WPCs. It is a well-established fact that weathering (UV radiation and moisture) will affect both the wood and plastic components in WPCs. However, most of the studies and researches carried out on this matter are still not enough to fill in the knowledge gaps. Thus, this study investigates how various weight percentages of rice husk, recycled polypropylene, and homopolymer polypropylene, when incorporated together, could affect the mechanical properties and photodegradation of wood polymer composites, especially after being subjected to UV irradiation.

1.4 Objectives

1. To identify the optimum compositions that is made up of WPP, RH, and HPP to fabricate the WPC specimens.
2. To determine the mechanical properties of recycled polypropylene/rice husk composites and the effects of photodegradation on the composites by using Fourier Transform Infrared Radiation (FTIR) analysis and correlate the mechanical properties and photodegradation of the specimens after UV irradiation through the use of Optical Microscopy (OM).

1.5 Scope of work

The properties of the wood polymer composites, before and after being subjected to UV irradiation, were obtained by carrying out the tensile test, flexural test, FTIR

analysis, and morphological study of the fractured surface by using an Optical Microscope. The composites were made up of several compositions of waste polypropylene, neat homo polymer polypropylene, and rice husk. The scopes of works conducted for this thesis are as follows:

1. Various compositions of waste polypropylene rice husk pellets obtained from the industry were prepared by mixing with homopolymer polypropylene. The various percentages of homopolymer PP at between 10 to 30% were added to act as additional reinforcements for the WPC pellets.
2. The mixtures of WPC pellets and homopolymer PP material were melt blended by using an injection molding process. The setting temperature and control parameter were predetermined.
3. The specimens were subjected to UV irradiation based on the ASTM D 4587-11: 2019 protocol and by using the weather meter machine. The UV irradiation processes were performed for 1000, 2000, 3000, 4000, 5000, and 6000 hours to study the effects of weathering on the composites.
4. Several tests were conducted to determine the mechanical properties of the WPC samples before and after going through the UV irradiation. The test included:
 - (a) Tensile test that was conducted according to ISO 527-2: 2012.
 - (b) A flexural test was carried out based on ISO 178: 2019.
5. Fourier Transform Infrared Radiation (FTIR) analysis was used to study the relationship between mechanical properties of WPC samples and photodegradation after being subjected to accelerated weathering.
6. The morphology of the fractured surface of specimens was studied by using Optical Microscope (OM). This analysis is essential to study the compatibility of the mixed materials further and to observe the dispersal of rice husk.

1.6 Significance of research

The significance of this research includes providing further insights into how different compositions of rice husk, recycled polypropylene, and homopolymer propylene may affect the mechanical properties of wood polymer composites. Apart from that, this research shows the photodegradation of the wood polymer composites, specifically in terms of the effects of the carbonyl index on the mechanical properties. The general-purpose grade of polypropylene homopolymer is the most widely utilized material in a wide range of industries. It contains only a propylene monomer in a semi-crystalline solid form. The main applications of this material include packaging, textiles, healthcare, pipes, automotive, electrical applications, and many more. Even though polypropylene is easily among the most popular plastic packaging materials in the world, but only around 1% of them are recycled, meaning that most of the polypropylene products are headed for the landfill, thus causing severe environmental issues.

The findings of this research could contribute to better management of plastic wastes, as well as widen the applications of recycle-able plastic wastes. Plastic wastes can be recycled to produce different types of products, including clothing fibers, industrial fibers, food containers, dishware and etc. This helps to reduce the use of virgin plastics that are made from non-renewable fossil oil. Hence, recycle-able plastic wastes are more environmentally sustainable and can be reclaimed from the waste streams; in addition, they can be easily recycled with minimal effort. Recycling and reusing plastic and other wastes are not only ecologically important but also economically crucial, as the cost of discarding wastes has increased, and this trend is expected to continue in the future.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Plastics are considered a necessity due to their widespread application in various industries, including automotive, electronics, buildings, and constructions sectors. About 90% of the demands for plastics demand are on five main commodities, namely polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polystyrene (PS), and polyethylene terephthalate (PET). However, the increased usage of this material is igniting concerns with environmental issues. Since plastic wastes are non-biodegradable, recycling this material for extra usage is now getting more attention as a solution to the increasing amount of municipal solid wastes. Figure 2.1 shows plastics demand distribution by resin type 2019.

PLASTICS DEMAND DISTRIBUTION BY RESIN TYPE 2019

Data for EU28+NO/CH.

SOURCE: PlasticsEurope
Market Research Group
(PEMRG) and Conversio
Market & Strategy GmbH

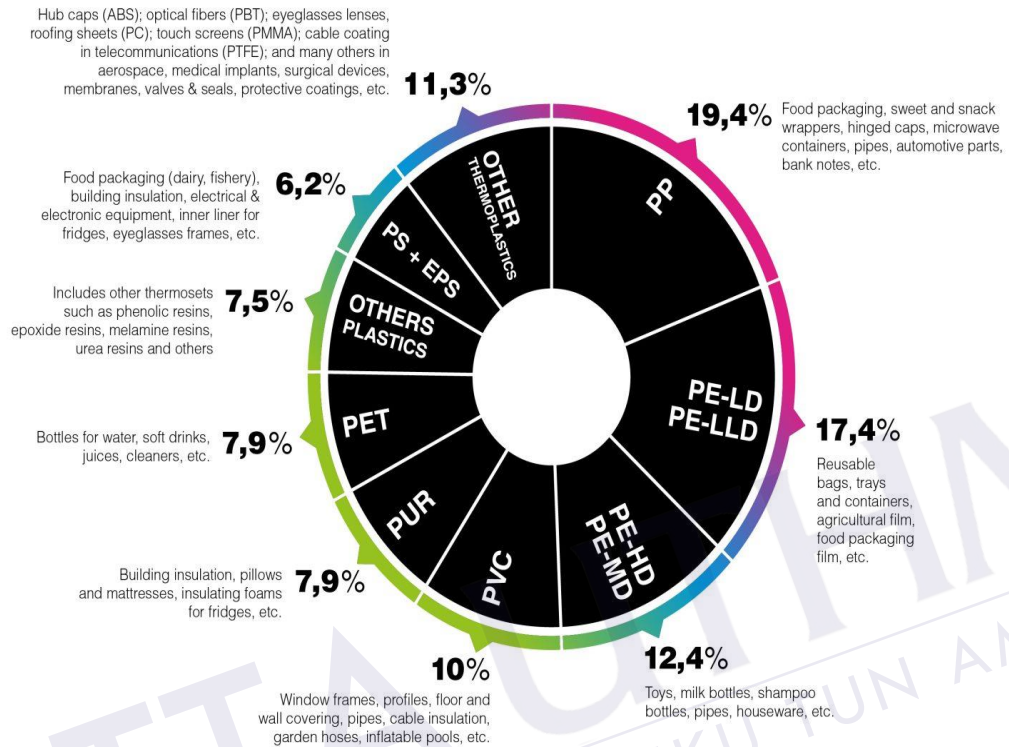


Figure 2.1: Distribution of plastic demand by resin type 2019

(Plastic: demand, types and destinations of use – In the world and in FITT, 2022)

2.1.1 History of wood polymer composites

Wood polymer composite is a composite that consists of wood and thermoset or thermoplastic. This material is still considered a new material, even though it is quite well known, with quite a number of researches have been conducting investigations on this material in the past several decades. However, it has only recently caught the attention of the wood-plastic industry worldwide.

Inorganic fillers and reinforcements were mostly used in the composite during the earlier practice, before the wood materials were used as a substitution, followed by recycled wood chips or wood flour. Wood substitution exhibits several advantages,

such as being lighter, less abrasive, renewable, and lower in cost. More importantly, this substitution improves the stiffness and dimensional stability, as well as with the minimal increase in weight. The usage of wood-plastic composites was limited before 1980 (Clemons, 2000), as these wood and plastic industries were not familiar and interrelated with each other. Furthermore, the lack of usage was also caused by very few materials and equipment suppliers, in addition to different scales of various process materials.

Wood polymer composite was first studied in the industry as an automotive interior by the American Woodstock company in 1983. Meanwhile, Italian extrusion technology was used to produce the WPC panel substrates. That technology produced polypropylene with approximately 50% of wood flour flat sheets by forming the materials into various shapes.

Later, in the early 1990s, Advanced Environmental Recycling Technology (AERT), together with a division from Mobil Chemical Company (Trex), started manufacturing solid wood polymer composites that were consisted of approximately 50% of wood fiber in polyethylene. These materials were produced into deck boards, landscape timbers, picnic tables, and industrial flooring. Today, the decking market is considered the leading and fastest-growing WPC market. Now, the extruded WPC profiles can be manufactured and shaped straight into their final products without having to go through the milling or further forming processes. Since then, several companies have begun to venture into the manufacturing of WPC products, and this eventually led to market expansion.

The market grew larger when several companies in the United States started to provide feed stock made from wood or other natural fibers and plastic (WPC compound) in pellet form to the processors who were not capable or did not want to blend their own material. Consequently, the WPC industry increased radically with the rapid development of technology and the increase of players in the market. In 1991, the first International Conference on Wood-fiber Plastic Composites was held in Wisconsin, United States. In this conference, ideas were being discussed, and cooperation took place between both the researchers and industrial representatives.

In conclusion, the utilization of wood-plastic composite as a substitute for plastic and steel components in the construction field is expected to increase in market growth due to a higher demand for low-cost and more environment-friendly materials. Apart from plastic wastes, the use of agricultural by-products, such as husks, straw, and

fiber, is increasing in the wood polymer composite industries. However, similar to plastic wastes, the difficulties with agro wastes include decomposition, trouble with digestion, and low nutritional values for animals. For example, lignocelluloses, a complex mixture of lignin and cellulose that is present in the cells of woody plants, could function as reinforcements in WPCs. The major setback of this application is the poor compatibility with plastics since lignin by-products are hydrophobic in nature, hence resulting in low mechanical properties. However, agro wastes are much more environmental-friendly, as compared to inorganic materials that are mostly residues and toxic by-products during the manufacturing processes.

The consumption of virgin plastics and environmental impacts can be reduced by reutilizing post-consumed polymeric materials. Recycling can be easily done on single polymer plastics that come from petroleum. This only requires a systematic and efficient process that includes collection, separation, recycling system, and addition of energy. The effects of fiber content, matrix type, and interfacial bonding on the tensile and flexural properties of composite materials were studied by Jayaraman and Bhattacharya (2004). The composite materials consisted of waste plastics obtained from high-density polyethylene wastes, and medium-density fibers were produced from melt blending and injection molding. It was found that the mechanical properties of these composites at room temperature and humidity was found to be dependent on the fibers' content, mechanical properties of the waste plastics used, and the presence of a suitable coupling agent. It was also found that the tensile and flexural modulus of the composites increased with increasing fiber content.

The use of agro wastes and natural fibers in the composites is increasingly getting the attention of fellow academicians and industrialists, due to certain properties, such as improved mechanical strength, the barrier against water and oxygen, dimensional stability, thermal and wear resistance, and other excellent qualities. In addition, due to its low cost, agro wastes fiber-reinforced plastic composites are receiving significant attention in the automobile and building industries. Beneficial properties, such as renewability, biodegradability, low specific gravity, availability, great strength, and non-abrasiveness, are making agro waste plastic composites preferable in various practical applications. As a highlight, highly rich lignocellulose materials are one of the under-utilized agro-wastes.

According to Mehdi et al., 2017, varying the percentage of sand dune and

recycled high-density polythene mixtures in designing roof tile caused the density to decrease alongside the increasing ratio of plastic waste. The findings also indicated that the breaking load values of all-polymer mix tend to increase below the values of the mixture reference and the standard, with increasing the amount of high-density polyethylene (HDPE), as shown. This is logical, as when the ratio of HDPE as binder increases, this gives the material enough bond strength that then leads to an increase in the breaking load. It was also discovered that the impermeability test of the roof tile mix showed better results, as compared to the conventional clay roof tile.

Polymer and plastic composites are usually strengthened with fibers, fillers, particulates, powders and other matrix reinforcements for improved strength and stiffness. The fiber-reinforced composites are materials in which a fiber made of one material is embedded in another material. The most common fibres used as composite materials are glass, carbon, or aramid. Other fibres such as paper, wood or asbestos have also been used. Epoxy, vinyl ester or polyester thermosetting plastic, and phenol formaldehyde resins are among the common polymer used in composites.

2.2 Polypropylene

Polypropylene (PP) is widely used in various applications due to its good chemical resistance and weldability. PP is ideal for several packaging applications, car parts and outdoor furniture. PP has very high resistance to absorbing moisture and chemical compared to other wide range of bases and acids. Its resistance toward fatigue and impact strength is also high. On top of that, it is also tough, heat-resistant, and has ability to retain its shape after a lot of torsion, bending, or flexing. All of these characters make PP more favorable compared to other plastics because it is considered safe for humans. Unlike other plastics that contain bisphenol A (BPA) or other harmful chemicals, PP does not negatively affect human bodies and environment.

According to Raymond et al., 1989, Polypropylene (PP) was first discovered in 1954 by Giulio Natta, followed by the polymerization of propylene monomer (Figure 2.2) by Karl Ziegler, also in 1954. A new process for synthesizing polymers to produce a lot of common plastics, including HDPE and PP, was discovered by Ziegler and Natta (1954). PP possess excellent chemical resistance and can be

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