MECHANICAL AND THERMAL PROPERTIES OF HYBRID RICE HUSK AND COCO PEAT FILLED ACRYLONITRILE BUTADIENE STYRENE POLYMER

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DEDICATION

Every challenging works need strength from people around me especially from my husband, Kapt Mohd Izzuddin Bin Khamarolzaman for always supports and being a backbone for me. My kids Nur Afrina Irdeena and Muhammad Afeef, this proposal dedicated for encouragement to keep learning.

My humble effort I dedicate to my parent, siblings and family in law. Thank you for all the prayers and love. This thesis represent appreciation to all of you.

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ABSTRACT

The utilization of natural waste material such as rice husk (RH) and coco peat (CP) that can fill a thermoplastic matrix to produce low-cost green composites are interesting to be studied. Weak properties that exist in rice husk and coco peat can be improved through hybridization methods which can reduce deficiencies in both fillers. In this study, the effectiveness of these hybridized biocomposites was evaluated based on the following physical, mechanical and thermal properties. Initially, the samples were fabricated using a combination of melt blend mixer and injection moulding techniques. The RH and CP fillers were incorporated with the acrylonitrile butadiene styrene (ABS) matrix in different weight fractions commencing with 0% RH - 5% CP, 5% RH - 15% CP, 10% RH – 10% CP, 15% RH – 5% CP and 20% RH – 0% CP respectively. It was found that an increment of rice husk weight ratio in hybrid fibre increased the density of samples. However, it reduced the kinetic water absorption of these biocomposites. Water absorption research exposed that the water uptake was increased at maximum of 20% with the increment compositions of coco peat filler. The 15% RH – 5% CP reinforced ABS exhibited an increment of 17.8% and 7.3% in tensile and flexural strength compared to the best result achieved from the monofibre RH. Result exhibited all rice husk and coco peat filled composites tend to be brittle fracture in manner. Observation on the tensile morphology surfaces using a scanning electron on microscope affirmed the above finding satisfactorily. The degradation period and thermal stability of the biocomposites indicated deterioration with the presence of rice husk and coco peat loading. Thus, it can be concluded that hybrid-natural waste reinforced ABS biocomposites can be utilized as a biodegradable material for engineering applications whereby excellent mechanical properties are paramount.

ABSTRAK

Penggunaan sisa buangan semulajadi seperti sekam padi (RH) dan sabut kelapa (CP) dapat diisi di dalam matrik plastik haba bagi menghasilkan komposit hijau yang berkos rendah menarik untuk diketengahkan. Sifat lemah yang terdapat di dalam RH dan CP dapat diperbaiki melalui kaedah hibridisasi, di mana boleh menutupi kekurangan dalam kedua-dua bahan pengisi. Dalam kajian ini, keberkesanan hibrid biokomposit dinilai dari ketahanan fizikal, mekanikal dan sifat haba. Awalnya, dengan sampel dihasilkan mengunakan teknik penggaul adunan lebur dan teknik pengacuan suntikan. Pengisi RH dan CP telah digabungkan dengan *acrylonitrile butadiene styrene* (ABS) matrik dengan pecahan berat yang berbeza iaitu 0% RH- 5% CP, 5% RH- 15% CP, 10% RH – 10% CP, 15% RH – 5% CP and 20% RH – 0% CP. Didapati bahawa semakin tinggi nisbah berat sekam padi di dalam hibrid gentian, maka semakin bertambah ketumpatan sampel. Walau bagaimanapun, ianya mengurangkan kinetik penyerapan air biokomposit tersebut. Kajian serapan air mendedahkan bahawa serapan air meningkat pada peratusan maksimum 20% dengan pertambahan komposisi pengisi sabut kelapa. 15% RH – 5% CP diperkuatkan ABS menunjukkan peningkatan sebanyak 17.8% dan 7.3% bagi kekuatan tegangan dan lenturan dibandingkan dengan keputusan yang terbaik diperolehi daripada monogentian RH. Hasil mempamerkan kesemua sekam padi dan sabut kelapa isian komposit cenderung kepada cara patah rapuh. Pemerhatian pada permukaan morfologi tegangan menggunakan mikroskop imbasan elektron mengesahkan penemuan di atas dengan jayanya. Tempoh penguraian dan kestabilan haba biokomposit menunjukkan kemerosotan dengan kehadiran beban sekam padi dan sabut kelapa. Oleh itu, ianya dapat disimpulkan bahawa sisa buangan hibrid diperkuatkan ABS boleh digunakan sebagai bahan biodegradasi untuk kegunaan kejuruteraan dengan sifat mekanikal yang cemerlang adalah menjadi keutamaan.

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LIST OF ABBREVIATIONS

ABS	-	Acrylonitrile Butadiene Styrene
RH	-	Rice Husk
CP	-	Coco Peat
FRP	-	Fiber Reinforced Polymer
DSC	-	Differential Scanning Calorimetry
TGA	-	Thermal Gravimetric Analysis
FESEM	-	Field Emission Scanning Electron Microscope
PLA	-	Polylactic Acid
HDPE	-	High Density Polyethylene
PP	-	Polypropelene
PVC	-	Polyvinyl Chloride
DTG	-	Derivative Thermogravimetric
ASTM	-	American Society Testing and Materials

LIST OF SYMBOLS

ρ	-	Density
m	-	Mass
V	-	Volume
W_a	-	Water Absorption
h	-	Specimen Thickness
M_m	-	Weight of Saturated Specimen

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CHAPTER 1

INTRODUCTION

1.1 Introduction

Currently, greener and biodegradable materials have attracted interest from researchers to be explored. The natural fibre is the famous material that has been focused on. Besides can be recycled, they have consisted of natural substances like cellulose, lignin, hemicellulose, pectin, and protein that offer outstanding performance, temperature stability, and high strength to weight ratio (Moosavi et al., 2020). According to 2014 data, the crops cultivated in Malaysia include rubber (39.67 per cent), oil palm (34.56 per cent), cocoa (6.75 per cent), rice (12.68 per cent), and coconut (6.34 per cent) (Tambichik et al., 2018). In general, Malaysia generates a million tons of agriculture residue a year, and the bulk of them are not fully utilized for further downstream operation. Figure 1.1 shows the production of agricultural waste in Malaysia in 2007 which was generated from the production of palm oil, rice, rubber, coconut, sugar cane waste, forest products, and municipal waste.

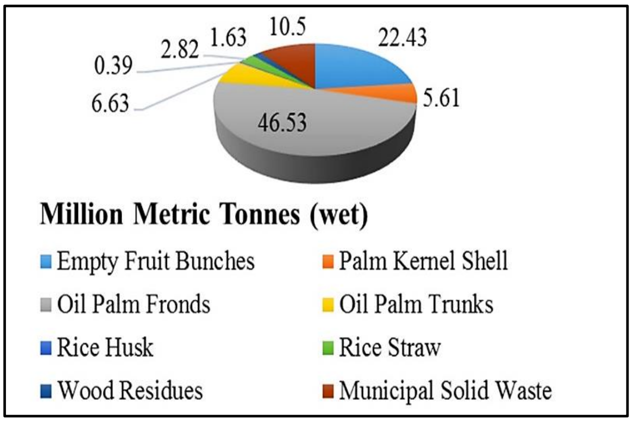

Figure 1.1 Production of agriculture waste in Malaysia (Mekhilef et al., 2011)

Malaysia as a leading producer of paddy in the world produced approximately 10 tonnes of paddy per hectare with the output of rice approximately 6,575,474.8 tonnes per year due to the emerging technological development in this sector (S. M. Shafie et al., 2012). According to Muda Agricultural Development Authority (MADA), it gained almost 0.48 million tonnes of rice husk (RH) in a year during the harvesting and milling process (Rosmiza et al., 2017). Table 1.1 shows the production of rice and waste from 2000 to 2012 in the project by the MADA agency. However, around 1.0% of waste has diversely been used in various applications including animal feed, compost, paper goods and mushroom growth media (MADA, 2014). Unfortunately, garbage burning remains the underlying cultural practice of disposal in Malaysia.

Table 1.1 Malaysia's total paddy and waste output during the season, 2000-2012 (Rosmiza et al., 2017)

Year	Planted area (hectares)	Paddy production (tonnes)	*Waste production (tonnes)
2000	698 702	2, 140 904	2,616 660
2001	673 634	2, 094 995	2,560 549
2002	678 544	2, 197 351	2,685 651
2003	671 820	2, 257 037	2,758 600
2004	676 310	2, 291 353	2,800 542
2005	666 781	2, 314 378	2,828 684
2005	676 034	2, 187 519	2,673 634
2007	676 111	2, 375 604	2,903 516
2008	656 602	2, 353 032	2,875 928
2009	674 928	2, 511 043	3,069 052
2010	677 884	2, 464 831	3,012 571
2011	687 940	2, 578 519	3,151 523
2012	684 545	2, 599 382	3,177 022

RH itself has several benefits, for example, it could be turned into fuel using the pyrolysis process. The highest energy content of the RH could be harnessed by this gasification technique to earn substantial combustion products to reduce the use of fossil fuels. RH also highly contains lignin and cellulose which are fitting in the use as activated carbon capture (Tiwari et al., 2017) (He. S et al., 2021) (Panda D et al, 2020). Wang et. al. (2020) modified the adsorbent X zeolite from RH for carbon dioxide (CO2) capture prepared using the consolidation of thermal and chemical activation. These modified zeolites enhanced the heat and gas absorption mechanisms during adsorption and desorption, which were suitable for long-term CO2 capture and separation from the industrial exhaust gas. Also, RH is an advanced material with a high composition of silica that can be used as a polishing and cleaning agent Tiwari et al., (2017). Furthermore, Okoya et. al. (2020) proposed to employ RH biochar as a catalyst in conventional water treatment for removing chlorpyrifos from pesticide polluted water.

Several studies reported the application of RH fibres in synthetic polymer for furniture and household appliances which could support the concept of reducing agriculture waste (Pratheep et al. 2020, Sadik et. al. 2021, Gupta et. al. 2020, Datta et. al.2020). Pratheep et al. (2020) also verified that high mechanical properties can be obtained by smaller RH size utilization as wood powder composite resultant from the reduction of void and cavities. This wood-plastic composite can be fully eligible as a decking and flooring fixture. Sadik et. al. (2021) also proved that the effect of nano-silica extracted from RH in wood plastic composite applications improved the water absorption and thickness swelling of composites. In addition, thermal stability was slightly enhanced as compared with neat composite. The utilization of RH fibres in composites as food packaging has got a lot of attention from researchers. Gupta et. al. (2020) synthesised the carboxymethyl cellulose (CMC) element from RH fibre to fabricate a biodegradable film. To improve the tensile strength and elongation of the film, the CMC was blended with glycerol and citric acid. Datta et al., (2020) also extracted RH fibre to produce crystalline starch phthalate. These thin films have excellent mechanical (tensile, tear, stiffness), optical (haze, transmittance), and biodegradation characteristics, making them ideal for biodegradable food packaging applications.

The coconut industry in Malaysia is considered as one of the new emerging crops that attracted a younger generation to be fostering and could bring a significant impact to the country's economy. Malaysian Agricultural Research and Development Institute (MARDI), reported that Malaysia is one of the top ten coconut growers in the world, although production decreased between 2014 and 2016., due can't compete with countries such as Thailand, Philippines and Indonesia in terms of price and volume (Hoe Kek Then et al., 2018). The area under coconut cultivation in Malaysia has gradually decreased from 120 000 ha in 2005 to 85 000 ha in 2016 reported by Tan Zhai Yun et al., (2019).

Furthermore, it is projected that this crop business provides a source of income for 80,000 people and supports a thriving small and medium-sized coconut-based industrial sector, with a positive trade surplus of around RM81.2 million in 2007. (http://www.anjungnet.mardi.gov.my, 2009). The reduction of area planted with coconut in Malaysia by 30 per cent within 10 years was mainly due to the conversion of coconut planting into oil palm or other crops. Figure 1.2 shows that coconut planting and production in 2005-2016 in Malaysia.

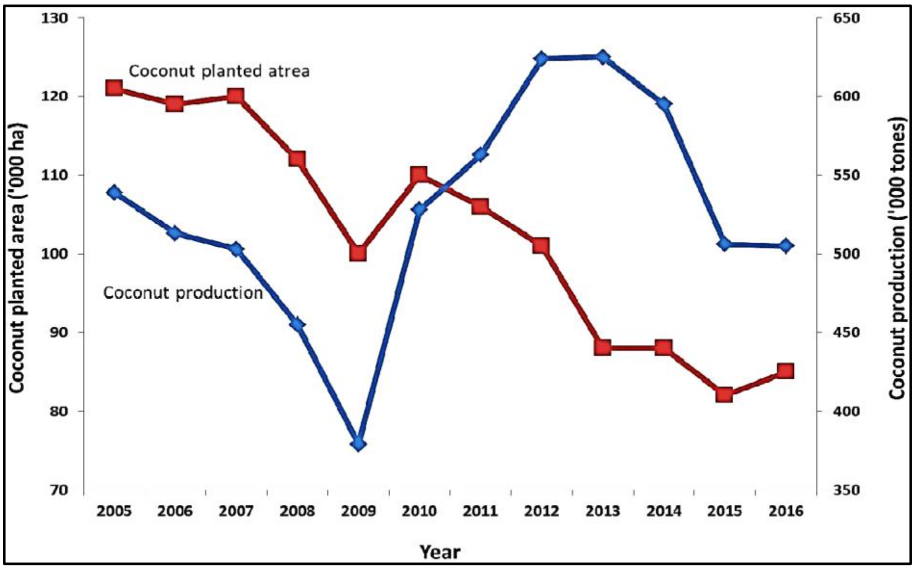

Figure 1.2 Malaysian coconut planting and production trends (2005-2016)

Coconut trash is a plentiful agricultural waste that also contributes to kitchen waste. The waste is produced by several components of the coconut head, including the husk, kernel, and flesh (inner part). Previously, such agro-waste was used as animal feed or organic fertiliser or was otherwise left to decay naturally in fields, dumped, or burned, but contemporary techniques are on-farm burning, burying, stockpiling and landfilling as reported by Sadh et al., (2018). Besides being used as animal feed or supplement, it may also be utilised as organic fertiliser via bioconversion.

Nowadays, composite materials have a wide range of applications in a variety of fields such as aerospace, automobile as well as sporting goods, due it has many advantageous properties such as high stiffness to weight ratio, specific strength, wear and corrosion resistance. For example, car manufacturing industries have begun to replace their conventional mild steel and aluminium parts with a composite structure to reduce the weight of the vehicle. Therefore, the utilizing of agro-waste materials as a recycled composite based product in the advanced application have been developed owing to their reuse, biodegradability and recycle (Wang et al., 2002).

Many researchers have concentrated their efforts on improving the properties of fibre-reinforced polymers. In general, two methods have been used in recent years, such as the hybridization of particulate filler in the matrices (Andrzejewski et al., 2020) (Ahmad S et al., 2019) and usage of different fibres (configuration, layering and varies of intraply design) in the woven composite laminates (Marino et al., 2021) (Margabandu et al., 2021) Many attempts have been made to improve the mechanical properties of acrylonitrile butadiene styrene (ABS) polymer through the filler hybridizing technique (Nikmatin et al., 2019) (Pramod et al., 2021). Nikmatin et al. (2019) evaluated the shock absorption, yield stress, frequency, and head injury criteria are among the mechanical characteristics of oil palm empty fruit reinforced ABS for helmets applications. Furthermore, Pramod et al (2021) mentioned that higher plastic deformation and irregular debris formation of betel nut reinforced ABS hybrid composite subjected to slide wear friction evaluation. These composites were not suitable for brake pads.

Based on past findings, even though extensive research has been explored the physical and mechanical properties of composites, very few have involved rice husk (RH) and coco peat hybrid biocomposites. Indeed, evaluating the hybridized RH and CP filler reinforced ABS composite on the physical and mechanical properties has not been conducted.

1.2 Problem Statement

Utilizing agriculture waste by using rice husk and coco peat reinforcing in thermoplastic polymer resin can produce low-cost green composites. Weak properties in rice husk and coco peat can be come across by hybridization techniques between both fillers. Higher-strength in rice husk has been used to encounter mechanical properties in coco peat. Coco peat shows that weak mechanical properties in terms of tensile and flexural strength. Therefore, hybrid rice husk in coco peat can enhance mechanical properties. Weak compatibility between the fiber and matrix and tends to drop the mechanical properties of the composites. Fiber agglomerations occurred when wettability between matrix and hybrid fiber as reported in Sun Z et al., (2018) Therefore, evaluating the maximum mechanical performance of the RH/CP reinforced ABS is interesting to disclose.

In addition, rice husk has a high content of silica and can be considered highly resistant to heat. of thermal stability of these hybrid composites was evaluated due to the hydrophilic character in coco peat. This filler significantly absorbs more water compare to rice husk and tend to change the density of the hybrid composites. In general, the water diffusion of natural fiber is higher than the polymer counterpart. In agro-waste/polymer composite, it happens via micro gaps between the polymer chains, flaws at the fiber-matrix interface and the matrix-cracking from the swelling effect during the curing process. The higher diffusion characteristic of the composite reduces the performance of the biocomposite. Here, the measurement of moisture absorption of these RH/CP reinforced ABS hybrid composites is required.

ABS has a limitation in thermal behavior. (Mohd et al., 2016) reported that the melting temperature of ABS is approximately 260°C. Therefore this behaviour can be turned by infusing an outstanding thermal resistance filler like RH. Predominantly, the thermal stability, endotherms, exotherms, weight loss on heating and cooling rate of new material are changed. The thermal stability of RH/CP of ABS biocomposites needs to be characterised through analysis of decomposition patterns, degradation mechanisms, and thermal reactions kinetics.

Besides that, from the above matter hybridization techniques can transfer the waste material into more valuable for various purposes and functional products. It also turns waste to perform new material. This technique also can reduce the usage amount of polymer and offer better performance.

1.3 Objectives of the Study

This study aims to measure the physical, mechanical and thermal degradation performance of the fully recycled RH/CP reinforced ABS biocomposite. The objectives of this research are as follows:-

i. To evaluate density and moisture absorption of RH/CP reinforced ABS biocomposite.
ii. To investigate the mechanical properties of RH/CP reinforced ABS biocomposite.
iii. To examine the thermal stability of RH/CP reinforced ABS biocomposite.

1.4 Significance of the Study

The increasing demand for eco-friendly materials and a new rule of environmental regulation toward increasing the depletion rate have forced a widening interest in the field of agro-waste biocomposite structures. The utilisation of agro-waste in advanced composite material tends to give a potential benefit to humans and sustainable green technology.

1.5 Scope of the Study

In this research, Rh and CP were used as a filler while ABS was a matrix. Five predetermined weight percentage ratios between RH and CP filler were used namely 20RH/0CP, 15RH/5CP, 10RH/10CP, 5RH/15CP and 0RH/20CP. Composites were fabricated used a twin-screw extruder, a chopping machine to get a smaller size and injection moulding to get samples.

Initially, the density and moisture absorption of the composites were evaluated. Then, the mechanical properties of this biocomposite including tensile, flexural and impact tests were examined. To follow suit, surface characterization of the composites was also obtained. Finally, the thermal degradation of these samples was measured. Five replicated samples were tested for each experimental work.

1.6 Content of the Thesis

This thesis consists of five main chapters where Chapter 1 introduced the hybrid fiber; rice husk and coco peat used as a filler, current situation of agriculture waste and hybrid composites applications. Other chapters are:

(a) Chapter 2 in the literature review covered cellulose base natural fiber, crop waste residue as filler, properties of rice and coco peat, hybridization, thermal stability and ABS as a polymer matrix.
(b) Chapter 3 mentioned the flowchart of the research, sample fabrication using injection moulding, testing on physical, mechanical and thermal properties.
(c) Chapter 4 discussed physical findings such as density and moisture absorption, the highest mechanical properties from testing mention in Chapter 3 and thermogravimetric analysis.
(d) Chapter 5 concluded the physical, mechanical and thermal properties of hybrid composite and recommendations for future work.

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APPENDIX A

LIST OF PUBLICATION

Journal with Impact Factor

1. **Norhasnan, N. H. A**., Hassan, M. Z., Nor, A. F. M., Zaki, S. A., Dolah, R., Jamaludin, K. R., & Aziz, S. A. A. (2021). Physicomechanical Properties of Rice Husk/Coco Peat Reinforced Acrylonitrile Butadiene Styrene Blend Composites. *Polymers*, *13*(7), 1171. **(Q1, IF: 3.426)**