UNIVERSITI PUTRA MALAYSIA

EFFECTS OF FIBRE LOADING AND NANO-SILICA ON PHYSICAL, MECHANICAL AND THERMAL PROPERTIES OF BAGASSE FIBREFILLED RECYCLE HIGH-DENSITY POLYETHYLENE

QAMARIAH NORHIDAYAH BINTI SALLEH

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By

QAMARIAH NORHIDAYAH BINTI SALLEH

Thesis Submitted to School of Graduates Study, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

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DEDICATION

This thesis is dedicated to special person in my life who taught me to trust in Allah, believe in handwork, give me the encouragement to believe in myself and support me all the way.

My late father, Salleh Bin Nik Mat
My stronger mother, Puan Atikah Binti Abdullah
My Family members

I hope I have made all of you proud
Thank you very much
Thanks *Allah s.w.t. Alhamdulillah*

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

EFFECTS OF FIBRE LOADING AND NANO-SILICA ON PHYSICAL, MECHANICAL AND THERMAL PROPERTIES OF BAGASSE FIBRE-FILLED RECYCLE HIGH-DENSITY POLYETHYLENE

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November 2019

Chairman : Adlin Sabrina binti Muhammad Roseley, PhD
Faculty : Forestry and Environment

Fibre-reinforced composite (FRCs) is an attractive natural fibre-based material to many applications due to its desirable properties including high strength-to-weight and stiffness-to-weight ratios, and corrosion resistance. In recent years, growing interest in FRCs has promoted an increase in engineering application such as aerospace, defence, automotive, construction, marine and oil and gas. As referred above, FRC is made of thermoplastic polymer reinforced with natural fibre to form into board. Natural material in form of fibres or flour is added into polymer matrix such as recycled high-density polyethylene (rHDPE) to form FRC. The composite made from wood and natural fibres are environmentally sound due to its biodegradable properties. Synthetic filler such as nano-SiO_2 is often used in FRC to enhance its properties. Since, FRC has a high potential to be alternative material in various industries, the better knowledge on physical, mechanical and thermal properties are the main concern in this study. Hence, the aim of the research was to examine the effect of bagasse fibre loading and nano-SiO_2 on the physical and mechanical properties of bagasse/rHDPE composite. This study also investigates on the thermal properties of bagasse/rHDPE composite in response to different fibre loading and nano-SiO_2 content via dynamic mechanical analysis. For this purpose, bagasse fibre in 0.5-0.9mm and 1.0-1.4mm particle size and various weight ratios (30 wt%, 50 wt% and 70 wt%) were mixed with rHDPE. In order to increase the interfacial adhesion between the filler and the matrix, MAPE was used as coupling agent. Nano-SiO_2 with weight ratio of 0, 2, 4wt% were utilized to enhance the composite properties. The physical properties (water absorption and thickness swelling) and mechanical properties (tensile, flexural and impact strength) were carried out on the samples based on ASTM standards. To determine the thermal properties of composite, DMA test was carried out. The mechanical properties

of BF/rHDPE composite shown an improvement with the addition of 30 wt%, 50 wt% and 70 wt% of fibre content. The study finds that composite with 70 wt% had the highest value of modulus of rupture (MOR), modulus of elasticity (MOE), tensile and impact strength compared to 30 wt% and 50 wt% fibre loadings. Modification of the composite using nano-SiO_2 filler has showed that composite with 4 wt% performed significantly better than 0 wt% and 2 wt% in tensile, flexural and impact strength properties. This study also indicates that water absorption (WA) and thickness swelling (TS) of BF/rHDPE composite increases with fibre loadings. Similar trends were observed with BF/rHDPE composite of larger particle size. The result for dynamic mechanical analysis (DMA) has indicated an increase of storage modulus (E'), loss modulus (E") and loss factor (tan δ) with the incorporation of short fibre (0.5-0.9mm) at 70 wt% fibre loading and 4 wt% of nano-SiO_2. Overall, BF/rHDPE composite produced with 70 wt% BF and 4 wt% of nano-SiO_2 had the best improvement in terms of physical, mechanical and thermal properties.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

KESAN KANDUNGAN SERAT DAN NANO-SILICA PADA SIFAT-SIFAT FIZIKAL, MEKANIKAL DAN TERMA KOMPOSIT DARIPADA SERAT TEBU BERPENGISI DENGAN KITAR SEMULA POLIETILENA BERKETUMPATAN TINGGI

Oleh

QAMARIAH NORHIDAYAH BINTI SALLEH

November 2019

Pengerusi : Adlin Sabrina binti Muhammad Roseley, PhD
Fakulti : Perhutanan dan Alam Sekitar

Komposit bertetulang serat (FRCs) adalah bahan berasaskan serat semulajadi yang menarik kepada banyak aplikasi kerana sifatnya yang diingini termasuk kekuatan yang tinggi untuk berat dan kekukuhan kepada nisbah berat, dan rintangan kakisan. Beberapa tahun kebelakangan ini, perkembangan yang memberangsangkan dalam FRC semakin meningkat dalam aplikasi kejuruteraan seperti aeroangkasa, pertahanan, automotif, pembinaan, laut, minyak dan gas. Seperti yang disebutkan di atas, FRC diperbuat daripada polimer termoplastik yang diperkuat dengan gentian semulajadi untuk membentuk papan. Bahan semulajadi dalam bentuk serat atau tepung ditambah ke dalam matriks polimer seperti polietilena berketumpatan tinggi (rHDPE) yang dikitar semula untuk membentuk FRC. Komposit yang diperbuat daripada kayu dan serat semulajadi adalah mesra alam disebabkan oleh sifat biodegradasinya. Pengisi sintetik seperti nano-SiO_2 sering digunakan di FRC untuk meningkatkan sifatnya. Memandangkan, FRC mempunyai potensi tinggi untuk menjadi bahan alternatif dalam pelbagai industri, pengetahuan yang lebih baik mengenai sifat fizikal, mekanikal dan terma adalah keutamaan dalam kajian ini. Oleh itu, tujuan penyelidikan ini adalah untuk mengkaji kesan pemuatan gentian bagasse dan nano-SiO_2 pada sifat fizikal dan mekanikal komposit bagasse / rHDPE. Kajian ini juga menyiasat sifat-sifat haba komposit bagasse / rHDPE sebagai tindak balas terhadap kandungan serat yang berbeza dan kandungan nano-SiO_2 melalui analisis mekanikal dinamik. Untuk tujuan ini, serat bagasse untuk saiz zarah 0.5-0.9mm dan 1.0-1.4mm dan pelbagai nisbah berat (30% berat, 50% berat dan 70% berat) dicampurkan dengan rHDPE. Untuk meningkatkan lekatan antara pengisi dan juga matriks, MAPE digunakan sebagai agen gandingan. Nano-SiO_2 dengan nisbah berat 0, 2, 4wt% digunakan untuk meningkatkan sifat komposit. Ciri-ciri fizikal (penyerapan air dan pembengkakan ketebalan) dan sifat-sifat mekanik (tegangan, lenturan dan kekuatan kesan) telah dijalankan ke atas sampel berdasarkan piawaian ASTM. Untuk menentukan sifat termal komposit, ujian DMA dijalankan. Sifat mekanikal komposit BF / rHDPE menunjukkan penambahbaikan dengan penambahan 30% berat, 50% berat dan 70% kandungan serat. Kajian mendapati bahawa komposit dengan 70% berat mempunyai nilai modulus pecah (MOR), modulus keanjalan (MOE), tegangan dan kekuatan impak berbanding dengan beban 30 wt% dan 50 wt%. Pengubahsuaian komposit dengan menggunakan pengisi nano-SiO_2 menunjukkan bahawa komposit dengan 4% berat lebih tinggi daripada 0% berat dan 2% berat pada sifat tegangan, lenturan dan kekuatan hentaman. Kajian ini juga menunjukkan bahawa penyerapan air (WA) dan ketebalan bengkak (TS) daripada BF / rHDPE komposit meningkat dengan meningkat nya kandungan serat. Trend yang sama diperhatikan kepada BF / rHDPE komposit dengan saiz zarah yang lebih besar. Keputusan untuk analisis mekanik dinamik (DMA) menunjukkan peningkatan modulus penyimpanan (E '), modulus kerugian (E) dan faktor kerugian (tan δ) dengan penggabungan serat pendek (0.5-0.9mm) dan 4% berat nano-SiO_2. Keseluruhannya, BF / rHDPE komposit yang dihasilkan dengan 70% berat BF dan 4% berat nano-SiO_2 mempunyai peningkatan yang terbaik dari segi sifat fizikal, mekanikal dan terma.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

Adlin Sabrina binti Muhammad Roseley, PhD
Senior Lecturer
Faculty of Forestry and Environment
Universiti Putra Malaysia
(Chairman)

Paridah Md. Tahir, PhD
Professor
Institute of Tropical Forestry and Forest Product
Universiti Putra Malaysia
(Member)

Sabiha binti Salim, PhD
Senior Lecturer
Faculty of Forestry and Environment
Universiti Putra Malaysia
(Member)

ZALILAH MOHD SHARIFF, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 11 March 2021

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I hereby confirm that:

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Signature: ______________________________

Name of Chairman of Supervisory Committee: Dr. Adlin Sabrina binti Muhammad Roseley

Signature: ______________________________

Name of Member of Supervisory Committee: Professor Dr. Paridah Md. Tahir

Signature: ______________________________

Name of Member of Supervisory Committee: Dr. Sabiha binti Salim

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LIST OF SYMBOLS

cm	Centimeter
g	Gram
g/cm^3	Gram Per Centimeter Cubic
g/mL	Gram Per Mililiter
Hz	Hertz
J	Joule
J/mm	Joule per millimeter
J/m	Joule per meter
kg	Kilogram
kg/m^3	Kilogram Per Meter Cubic
kN	Kilo Newton
mm	Milimeter
mm^2	Milimeter square
mm/min	Milimeter per minute
MPa	Mega Pascal
N	Newton
nM	Nanometer
Pa	Pascal
rpm	Revolutions per minute
SiO_2	Silica Dioxide
wt	Weight
wt%	Weight Percent
%	Percent
^{o}C	Celsius
$^{o}C/min$	Celsius per minute

LIST OF ABBREVIATIONS

AA	acrylic acid
ASTM	American Society for Testing and Materials
CBp	Carbonized
CCA	Chromate Copper Arsenate
CNTs	Nanotubes
DCP	Dicumyl Peroxide
DMA	Dynamic Mechanical Analysis
EBGMA	Ethylene/n-buty acrylate/glycidyl methacrylate
EMA	Ethylene-methyl acrylate
FTIR	Fourier Transform Infra-Red Spectroscopy
HA	Hydroxyapatite
HDPE	High Density Polyethylene
LDPE	Low Density Polyethylene
MASEBS	Maleated triblock copolymer stryrene-ethylene/butylene-stryrene
MAPE	Maleated Polyethylene
MCM-41	Mesoporous Silica
MOE	Modulus of Elasticity
MOR	Modulus of Rupture
MDPE	Medium Density Polyethylene
MWCnTs	Multi-wall Carbon Nanotubes
Nano-SiO_2	Nano-Silica
NAOH	Sodium Hydroxide
NC	nanoclay
PC	Polycarbonate

PET	Polyethylene terephthalate
PETr	Poly (ethylene terephthalate)
PLA	Polylactide
PP	Polypropylene
PS	Polystyrene
PVC	Poly Vinyl Chloride
rHDPE	Recycled High Density Polyethylene
SAN	ɣ- Methacryloyloxy Trimethyl Silane
SEM	Scanning Electron Microscope
TiO_2	Titanium Dioxide
TGA	Thermogravimetric analysis
TS	Thickness Swelling
UBP	Uncarbonized
USP	Unsaturated Polyester
vHDPE	Virgin High Density Polyethylene
WA	Water Absorption
WF	Wood Flour
WPC	Wood Plastic Composite
WPNC	Nanocomposite
XRD	X-Ray Diffraction

CHAPTER 1

INTRODUCTION

1.1 Background

The technology of fibre-reinforced composite (FRC) was born as a modern concept in Italy in the 1970s. FRC technology was popularized in North America in the 1990s and introduced to Singapore, India, Japan, Malaysia, and China by the early 21st century (Pritchard, 2004). The demand for fibre-reinforced composite (FRC) product has become rapidly increased in recent years. The popularity of FRCs are due to their high durability, low maintenance, and other insect attacks properties and resistance to termites. However, the composite has been limitedly used due to its high cost in production and low-strength instances.

The FRCs term may refer to any composite that contains any natural fibre resources and combines with thermoplastics or thermoset plastic as a binder. Thermoset may includes plastic materials such as epoxy, polyester, phenolic, polyurethane and polyimide and can be characterized by its inability to be melted once cured. On the contrary, the thermoplastic material can be re-melted repeatedly and the type of plastics may include polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC). FRCs can be found in various applications in building industry, automotive, furniture, and construction (Panthapulakkal *et al*., 2006).

High-density polyethylene (HDPE), polypropylene (PP), low-density polyethylene (LDPE/LLDPE), polyethylene terephthalate (PET), polystyrene (PS) and polyvinyl chloride (PVC) are the primary constituents of plastics in municipal solid waste (MSW). The blend of mixed waste plastics can be changed depending on the regional habits and season of a year. A statistic shown by the Department of National Solid Waste Management in 2011, the average composition of the waste collected from various industries were comprised of 36% plastic components, paper 35%, and food waste at only 6% of the total waste. The waste plastic or recycled HDPE obtained from the post-consumer milk bottles was fewer different from those of virgin resins and thus could be used for various applications. Hence, the recycled HDPE is cheaper than its virgin form. For instance, rHDPE pellets and flakes are 31-34% less expensive than the virgin HDPE (vHDPE).

In recent years, with the growth of environmental awareness, new legislation and ecological concerns, fibre-reinforced plastic composites have received increasing attention. The composites have many advantages including low energy consumption, lower cost, recyclability, and environmental friendliness. Fibre-reinforced composite is becoming a very common replacement for hardwoods in non-structural applications such as outdoor decking and furniture, landscaping, and cladding. The advantages of FRC such as rot resistance and lower maintenance cost have placed FRC's importance over the real timber and is considered as an environmentally friendly alternative. A large quantity of the fibre-reinforced composites on the market are produced from recycled plastics and sawdust/wood chip waste and it was promoted as an environmentally positive product. Nowadays, composite materials are being widely accepted in building construction, furniture, architecture material and recently used in the automotive industry (Smith *et al.,* 2006).

On the other hand, the main disadvantages of FRCs are an incompatibility between the hydrophilic natural fibres and hydrophobic polymers which resulted in non-uniform dispersion of fibres within the matrix and poor mechanical properties. In order to improve the affinity and adhesion between fibres and thermoplastic in production, two major components are used which is synthetic filler like nano-SiO_2 and coupling agent (MAPE) have been added (Kim *et al.,* 2006).

This study aims to explore the use of waste bagasse fibre and recycled high-density polyethylene for the production of the fibre-reinforced composites. The effect of the bagasse fibre content and nano-SiO_2 addition on the physical, mechanical and thermal properties were investigated.

1.2 Problem Statement

The composite industry has always looked into the alternative low-cost lignocellulosic source to decrease the manufacturing cost and increase the stiffness of the material. Natural fibre source such as sugarcane bagasse fibre, rice husk, cornstalks, and cereal straw has been recently used as an alternative to synthetic fibre. Sugarcane bagasse waste is chose as a raw material because of its low fabricating costs and high-quality green end material. Similar to the use of polymer matrix, it can be observed from daily life due to the increasing uses of polymeric material. Thereby, the interest in recycled materials that developed from post-consumer polymers have gained bigger attention from the largest fraction of polymer waste that consists of polyolefin such as polyethylene (PE), polyethylene terephthalate (PET), polyvinyl chloride (PVC), low-density polyethylene (LDPE/LLDPE), polystyrene (PS), polypropylene (PP), polylactide (PLA), polycarbonate (PC).

However, there are certain disadvantages of the composite reinforcement using natural fibres. The problem arises from poor compatibility between fibre and matrix along with their relatively high moisture absorption. Therefore, natural fibre modifications are considered as a solution to improve their adhesion by using different matrices, compatibilizer, and synthetic filler. The stiffness and an exemplary strength could be achieved through a stronger bonding between fibre and matrix resin and by the incorporation of rigid fillers.

To overcome this problem, this study is conducted in order to develop the composite from recycled material such as sugarcane waste (bagasse fibre) and recycled high-density polyethylene (rHDPE) modified with nano-SiO_2 powder. Thus, the combination of natural fibre and polymer is known as fibre reinforced composites (FRCs). This study utilizes nano-SiO_2 as filler due to high strength, high aspect ratio platelets and stiffness, very strong absorbability and high gas barrier quality. By intensively implementing this study on the physical aspect, mechanical and thermal properties of FRCs, these could help to improve the FRCSs on market space.

1.3 Research Objective

This study generally aims to determine the influence of bagasse fibre, polymer matrix, a coupling agent (MAPE) and synthetic filler (nano-SiO_2) on the physical and mechanical properties of fibre-reinforced composites. The evaluation of the composite can be represented through three objectives:

a. To examine the effect of bagasse fibre content and nano-SiO_2 on the physical of fibre-reinforced composites.
b. To investigate the mechanical properties of fibre-reinforced composites with the effect of different fibre loading and nano-SiO_2 content.
c. To evaluate the thermal properties of fibre-reinforced composites in response to different fibre loading and nano-SiO_2 content via dynamic mechanical analysis.

1.4 Scope of Study

The study focuses on the fibre content, particle size and nano-SiO_2 on the properties of the fibre-reinforced composites via physical, mechanical and thermal characterization. The testing was conducted according to the ASTM standard requirement. Sugarcane bagasse and recycled HDPE were used as reinforcement in the composite while nano-SiO_2 is added as a filler to improve the mechanical properties of fibre-reinforced composites. Maleated polyethylene (MAPE) was used as a compatibilizer agent to improve the interfacial adhesion between fibre and resin matrix.

1.5 Significance of Study

Nowadays, fibre-reinforced composites (FRCs) have become increasingly important in the construction industry and for home improvement, gardening, municipal, automobile and other application such as pallet construction. Theoretically, FRCs are made from plant fibre and thermoplastic materials. These materials can be obtained from waste plastic and scrap timber sources. The result of the production and with the support of FRCs could “turn the wastes into something useful”. However, to compare with the actual woods, several disadvantages are associated with FRCs, including on their impact and tensile strength. Due to that, this study should be processed in future development. These research findings can be a guideline to other researchers to produce any new conceivable ideas through the application of natural fibre (bagasse) reinforced by nano-SiO_2.

1.6 Thesis Outline

This thesis is primarily divided into five chapters. Chapter 1 indicate the background of this research. In addition, this chapter states the problem statement, objectives, the significance of the study and research limitations. Chapter 2 highlights the general literature review based on the related research areas of this study. Chapter 3 illustrates the phases in research methodology such as the preparation of material, testing procedure, and data collection. Chapter 4 discusses the additional results of this research including the analysis and presentation of the data and the correlation between the findings. Chapter 5 states the overall conclusion from the whole study as well as the future recommendations for further improvement of this work.

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BIODATA OF STUDENT

Qamariah NorHidayah binti Salleh was born 28 April 1988, in Kedai Mulong, Kelantan, Malaysia. She completed her secondary school in Sekolah Menengah Teknik Pengkalan Chepa in the year 2005. Then, she continued her study in Civil Engineering at Politechnic Kota Bharu in Year 2007. Later, she pursued her studies for Degree level in Faculty of Forestry, Universiti Putra Malaysia. She obtained her Bachelor's Science in Wood Technology in 2015. Currently, she continued her studies for Master level at Faculty of Forestry, University Putra Malaysia in the field Bio resource and Technology.