PREPARATION AND CHARACTERIZATION OF RECYCLED HIGH-DENSITY POLYETHYLENE HYBRID COMPOSITES FOR WEATHERING AND THERMAL AGING PERFORMANCE

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DOCTOR OF PHILOSOPHY

2025

Faculty of Industrial and Manufacturing Technology and Engineering

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A thesis submitted
in fulfillment of the requirements for the degree of
Doctor of Philosophy

Faculty of Industrial and Manufacturing Technology and Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2025

DECLARATION

I declare that this thesis entitled "Preparation and Characterization of Recycled High-Density Polyethylene Hybrid Composites for Weathering and Thermal Aging Performance "is the result of my own research except as cited in the references. The thesis has not been accepted to any degree and is not concurrently submitted in candidature of any other degree.

Signature :..

Name :.Ts. THINAKARAN A/L NARAYANAN

Date :.. 25.07.2025 ....

APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the award of Doctor of Philosophy.

Signature : ....

Supervisor Name : PM. Ir. Ts. Dr. JEEFFERIE BIN ABD. RAZAK

Date : 25.07.2025

DEDICATION

To my adored wife Thilagalakshmi Manogaran and wonderful children's, Daakshavarthan Thinakaran, Roshan Thinakaran, Tharunraj Thinakaran and Vahrun Thinakaran whose tireless efforts and unconditional love have been the pillars of my success. You have carried the weight of our family while I pursued this rigorous path, and for that, I am eternally grateful. This thesis is a tribute to your strength and sacrifice. It is a declaration of my love and appreciation, and a promise to continue honoring the legacy we build together.

ABSTRACT

This research focuses on the preparation and characterization of recycled high-density polyethylene (rHDPE)/calcium carbonate ($CaCO_3$) hybrid composites reinforced with fiberglass (FG) and waste rubber (WR), targeting structural applications in agricultural and outdoor environments. Objective 1 optimized the single-screw extrusion process using a two-level full factorial Design of Experiments (DOE), identifying optimal conditions as 260°C mixing temperature, 20 rpm screw speed, and an 80:20 wt% ratio of rHDPE to $CaCO_3$, achieving improved mechanical properties, tensile strength of 18.89 MPa, flexural strength of 48.89 MPa, and compressive strength of 10.43 MPa. Objective 2 evaluated the effects of hybrid fillers on mechanical, thermal, flammability, and physical properties, revealing that FG significantly enhanced structural strength with a tensile strength of 12.6 MPa, while WR improved impact resistance and toughness. UL 94 flammability tests showed that FG 7030 achieved the shortest burn times (17.24 s for 25 mm and 110.78 s for 100 mm), and Limiting Oxygen Index (LOI) values increased from 18.5% for pure rHDPE/$CaCO_3$ to 37.78% for FG 6040, indicating enhanced flame retardancy. Thermogravimetric analysis (TGA) confirmed improved thermal stability with a decomposition onset temperature of 481.3°C, while DSC results showed good crystallinity retention, confirming minimal thermal degradation. FTIR analysis revealed chemical interactions between the matrix and fillers through carbonyl (C=O) and silicate (Si–O–Si) bonding, which improved interfacial adhesion. XRD patterns confirmed the preservation of crystallinity in the rHDPE matrix and effective integration of fillers without disrupting the lattice structure. Objective 3 assessed the outdoor performance under natural weathering and accelerated thermal conditions, where the selected formulation (70% rHDPE/$CaCO_3$, 21% FG, 9% WR) maintained its mechanical and thermal stability after 90 days of exposure, with only minor degradation observed in tensile strength (from 12.8 MPa to 12.6 MPa). FESEM analysis confirmed consistent filler dispersion and strong interfacial bonding. Overall, hybrid filler reinforcement significantly enhances the mechanical, thermal, and environmental resilience of rHDPE-based composites. By effectively utilizing recycled and industrial waste materials, this study offers a sustainable, high-performance alternative for structural use in construction, agriculture, and automotive industries.

Penyediaan dan Pencirian Komposit Hibrid Kitar Semula Polietilena Berketumpatan Tinggi untuk Prestasi Luluhawa dan Penuaan Haba

ABSTRAK

Penyelidikan ini memberi tumpuan kepada penyediaan dan pencirian komposit hibrid kitar semula polietilena berketumpatan tinggi (rHDPE)/kalsium karbonat ($CaCO_3$) yang diperkukuh dengan gentian kaca (FG) dan getah sisa (WR), menyasarkan aplikasi struktur dalam persekitaran pertanian dan penggunaan luar. Objektif 1 mengoptimumkan proses penyemperitan skru tunggal menggunakan reka bentuk eksperimen (DOE) faktorial penuh dua peringkat, mengenal pasti keadaan optimum sebagai suhu pencampuran 260°C, kelajuan skru 20 rpm, dan nisbah 80:20% wt% rHDPE kepada $CaCO_3$, mencapai sifat mekanikal yang lebih baik, kekuatan tegangan 18.89 MPa, kekuatan lentur 48.89 MPa, dan kekuatan mampatan 10.43 MPa. Objektif 2 menilai kesan pengisi hibrid terhadap sifat mekanikal, haba, mudah terbakar dan fizikal, mendedahkan bahawa FG meningkatkan kekuatan struktur dengan ketara dengan kekuatan tegangan 12.6 MPa, manakala WR meningkatkan rintangan hentaman dan keliatan. Ujian mudah terbakar UL 94 menunjukkan bahawa FG 7030 mencapai masa pembakaran terpendek (17.24 s untuk 25 mm dan 110.78 s untuk 100 mm), dan nilai Indeks Oksigen Mengehadkan (LOI) meningkat daripada 18.5% untuk rHDPE/$CaCO_3$ tulen kepada 37.78% untuk FG 6040, menunjukkan kalis api yang dipertingkatkan. Analisis termogravimetri (TGA) mengesahkan kestabilan haba yang lebih baik dengan suhu permulaan penguraian 481.3°C, manakala keputusan DSC menunjukkan pengekalan penghabluran yang baik, mengesahkan degradasi haba yang minimum. Analisis FTIR mendedahkan interaksi kimia antara matriks dan pengisi melalui ikatan karbonil (C=O) dan silikat (Si–O–Si), yang meningkatkan lekatan antara muka. Corak XRD mengesahkan pemeliharaan kehabluran dalam matriks rHDPE dan penyepaduan pengisi yang berkesan tanpa mengganggu struktur kekisi. Objektif 3 menilai prestasi luar di bawah luluhawa semula jadi dan keadaan haba dipercepatkan, di mana formulasi yang dipilih (70% rHDPE/$CaCO_3$, 21% FG, 9% WR) mengekalkan kestabilan mekanikal dan termanya selepas 90 hari pendedahan, dengan hanya kemerosotan kecil yang diperhatikan dalam kekuatan tegangan (daripada 12.8 MPa kepada 12.6 MPa). Analisis FESEM mengesahkan penyebaran pengisi yang konsisten dan ikatan antara muka yang kuat. Secara keseluruhan, tetulang pengisi hibrid meningkatkan daya tahan mekanikal, haba dan alam sekitar komposit berasaskan rHDPE dengan ketara. Dengan menggunakan bahan buangan kitar semula dan industri dengan berkesan, kajian ini menawarkan alternatif yang mampan dan berprestasi tinggi untuk kegunaan struktur dalam industri pembinaan, pertanian dan automotif.

ACKNOWLEDGEMENT

First and foremost, I express my deepest gratitude to my respected supervisor, AP Ir. Ts. Dr. Jeefferie Bin Abd Razak, whose invaluable guidance, unwavering support, and profound wisdom have been the foundation of my research journey. His patience, insightful critiques, and constant encouragement have not only shaped this thesis but have also nurtured my growth as a researcher and scholar. I am truly blessed to have been under his mentorship.

I would like to extend my sincere appreciation to the examination panel, whose expertise and thoughtful feedback greatly contributed to the refinement of this work. To the Chairman of the Examination Committee, Profesor Madya Ts. Dr. Rostam Affendi bin Hamzah, thank you for your leadership and professionalism throughout the process. To the External Examiner, Profesor Madya Dr. Poppy Puspitasari, I am deeply grateful for your meticulous review, constructive comments, and academic generosity. To the Internal Examiner, Profesor Madya Ir. Ts. Dr. Mohd Yuhaizri bin Yaakob, thank you for your valuable insights and for upholding the highest standards of scholarly excellence.

My heartfelt thanks also go to the School of Graduate Studies, for your administrative support, guidance, and dedication in ensuring a smooth and meaningful academic journey. Your commitment to excellence has made a lasting difference in my experience as a postgraduate researcher. To my beloved family and cherished friends your understanding, patience, and unconditional love have been my anchor through every high and every challenge. In moments of doubt, your prayers and encouragement lifted my spirit and renewed my resolve. This achievement is as much yours as it is mine.

I am equally grateful to my dedicated colleagues at Institut Kemahiran Tinggi Belia Negara (IKTBN) Sepang and the Kementerian Belia dan Sukan (Ministry of Youth & Sports Malaysia). Your cooperation, motivation, and moral support have been a constant source of strength. Working alongside such committed and inspiring individuals has been both an honour and a privilege. This thesis is more than a culmination of research it reflects the collective effort, faith, and kindness of all who walked beside me, seen and unseen. To every person who believed in me, guided me, or simply stood by me in silence, I dedicate this milestone with profound gratitude and a heart full of humility.

Thank you, from the depths of my soul.

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LIST OF ABBREVIATIONS

UTeM	-	Universiti Teknikal Malaysia Melaka
FTKIP	-	Fakulti Teknologi dan Kejuruteraan Industri dan Pembuatan
IKTBN	-	Institut Kemahiran Tinggi Belia Negara
HDPE	-	High Density Polyethylene
rHDPE	-	Recycled High Density Polyethylene
$CaCO_3$	-	Calcium Carbonate
WR	-	Waste Rubber
FG	-	Fiberglass
FTIR	-	Fourier Transform Infrared
XRD	-	X-ray diffraction
DOE	-	Design of Experiment
FESEM	-	Field Emission Scanning Electron Microscope
DSC	-	Differential Scanning Calorimetry
TGA	-	Thermogravimetric analysis
RSM	-	Response surface methodology
WC	-	Waste Carpet
PVC	-	Polyvinyl Chloride
UV	-	Ultraviolet
HDT	-	Heat Deflection Temperature
CTE	-	Coefficient of Thermal Expansion
OPE	-	Oxidized Polyethylene
MICP	-	Microbial-Induced Calcium Carbonate Precipitation
ANOVA	-	Analysis of Variance
IM	-	Injection Molding
ASTM	-	American Standard Testing Method

UTM	Universal Testing Machine
UL 94	Underwriters Laboratories 94
LOI	Limiting Oxygen Index
BBD	Box–Behnken Design
HPC	Hybrid Polymer Composites
PMC	Polymer matrix composites
RH	Rice Husk
RTPW	recycled thermosetting polymer waste
FRP	Fibre Reinforced Polymer
GFRP	Glass Fibre Reinforced Polymer
rpm	Revolutions per Minute
wt. %	Weight Percentage
MPa	Mega Pascal
GYP	waste gypsum
SS	Soapstone
BRF	Banana Rachis fibers
WF	Wood flour
WSF	Walnut Shell Flour
RHF	Rice husk flour
DIY	Do it yourself
mm	Millimeter
min	Minute
kN	Kilo Newton
KJ	Kilo Joule
m^2	Square meter
m	Mass
v	Volume

LIST OF SYMBOLS

Ca^{2+}	-	calcium ions
°C	-	Degree Celsius
%	-	Percentage
et. al	-	et alia
g/cm^3	-	grams per cubic centimeter
W/m·K	-	Watts per meter-Kelvin
CO_3^{2-}	-	carbonate ions
H^+	-	hydrogen ions
σ	-	Sigma
α	-	Alpha
ϕ	-	volume fraction
wt.%	-	Weightage percentile
mm/min	-	millimeters per minute
:	-	Ratio
ρ	-	rho