UNIVERSITI TEKNIKAL MALAYSIA MELAKA

Faculty of Mechanical Engineering

FABRICATION AND CHARACTERISATION OF CYMBOPOGAN CITRATUS FIBRE REINFORCED THERMOPLASTIC CASSAVA STARCH/PALM WAX COMPOSITES

Zatil Hafila binti Kamaruddin

Doctor of Philosophy

2023

FABRICATION AND CHARACTERISATION OF CYMBOPOGAN CITRATUS FIBRE REINFORCED THERMOPLASTIC CASSAVA STARCH/PALM WAX COMPOSITES

ZATIL HAFILA BINTI KAMARUDDIN

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

Faculty of Mechanical Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2023

DECLARATION

I declare that this thesis entitled "Fabrication and Characterisation of Cymbopogan Citratus Fibre Reinforced Thermoplastic Cassava Starch/Palm wax Composites" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : ……………………………….…..

Name : Zatil Hafila Binti Kamaruddin

Date : 03/03/2023 ……………………………..….

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 : Dr. Ridhwan bin Jumaidin

Date : 03/03/2023

DEDICATION

To Al-Quran, the greatest source of knowledge

Bring me sheets of iron" - until, when he had leveled [them] between the two mountain walls, he said, "Blow [with bellows]," until when he had made it [like] fire, he said, "Bring me, that I may pour over it molten copper." (Al-Kahf:Verse 96)

and

To my beloved father and mother for their invaluable sacrifices, encouragements and support throughout my life

and

To my beloved husband for his love, patience and understanding

and

To my beloved son

and

To my awesome siblings

and

To all my supportive friends

ABSTRACT

Plastics manufactured from fossil fuels have made a substantial contribution to the environmental pollution that has been created by the accumulation of non-biodegradable trash, notably in the form of disposable products. In order to address this issue, renewable natural biopolymers emerge as an essential alternative to replace conventional plastic. Starch is one of the most widely available biopolymers and has been considered to suit many industrial needs owing to its renewability, abundant availability, biodegradability, and competitive price. Starch has also been found to have the ability to create rigid materials, notably thermoplastic starch. *Cymbopogan citratus* is a versatile plant that is regarded as a renewable source of natural fibre. However, pure thermoplastic starch also has several limitations, including low mechanical strength, long-term stability, and water resistance, limiting its potential applications. Meanwhile, palm wax is recognised as an excellent hydrophobic material because it has the potential to improve hydrophobicity of material. In order to improve the properties of native cassava starch (CS), blending starch matrix with hydrophobic material could enhance the biopolymer's performance. Apart from that, *Cymbopogan citratus* fibre (CCF) is a potential reinforcement for polymer composites. Hence, characterisations of the *Cymbopogan citratus* fibre were carried out to analyse its potential as a reinforcement material. Then, several modification methods were employed to enhance the properties of thermoplastic cassava starch (TPCS) i.e.; (1) blending TPCS with palm wax (2) reinforcement of TPCS/PW blend with *Cymbopogan citratus* fibre, and (3) alkali treatment the CCF fibre in the TPCS/PW blend. Consequently, TPCS/PW/CCF composite was successfully developed by using the hot pressing method. In terms of results, the findings showed that the mechanical properties of the material were improved following the incorporation of palm wax. The thermal properties of the material were slightly improved as the palm wax loading increased from 0 to 15 wt%. The development and characterisation of TPCS with the incorporation of 5 wt.% palm wax loading indicated the optimum strength of mechanical properties. Moreover, improvements in the mechanical properties of the TPCS/PW blends were evidenced after the incorporation of *Cymbopogan citratus* fibre. The results showed the improved mechanical properties of the TPCS/PW blend with CCF incorporation, with 50 wt.% CCF content yielded the maximum modulus and strength. It is also evident from the X-ray diffraction analysis (XRD) results that the crystallinity index of the composites was enhanced with the addition of CCF. In terms of thermal properties, CCF addition improved the material’s thermal stability, as shown by a higher-onset decomposition temperature and ash content. After soil burial for 2 and 4 weeks, the CCF incorporation into TPCS/PW slowed down the biodegradation of the composites. Lastly, the effect of alkali treatment on *Cymbopogan citratus* fibre into thermoplastic cassava starch/palm wax blends was evaluated. Obtained results indicated that the treated composite showed significant improvement in mechanical properties at 6% NaOH solution where the sample exhibited 19.9 MPa, 30.0 MPa, and 13.3 MPa for tensile, flexural, and impact strength, respectively. It is also evident that alkali treatment of the fibre has led to higher water resistance while soil burial results demonstrated slower biodegradability for the treated composites. Overall, the findings from this study demonstrated that TPCS modified by palm wax, *Cymbopogan citratus* fibre, and alkali-treated fibre has shown improved functional characteristics than the origin material. Hence, this study enhances the potential of thermoplastic cassava starch to be developed as alternative biodegradable material.

FABRIKASI DAN PENCIRIAN SERAT CYMBOPOGAN CITRATUS DIPERKUKUH DENGAN TERMOPLASTIK KANJI UBI KAYU/LILIN SAWIT KOMPOSIT

ABSTRAK

Plastik yang dihasilkan daripada bahan api fosil telah memberikan sumbangan yang besar kepada pencemaran alam sekitar yang telah dihasilkan oleh pengumpulan sampah tidak terbiodegradasi, terutamanya dalam bentuk produk pakai buang. Oleh itu, untuk mengatasi isu ini, biopolimer semula jadi yang boleh diperbaharui muncul sebagai alternatif penting untuk menggantikan plastik konvensional. Kanji adalah salah satu biopolimer yang tersedia secara meluas dan telah dianggap sesuai dengan banyak keperluan industri kerana boleh diperbaharui, ketersediaan yang banyak, kebolehbiodegradan, dan harga yang kompetitif. Kanji, juga didapati mempunyai keupayaan untuk mencipta bahan tegar, terutamanya kanji termoplastik. Cymbopogan citratus adalah tumbuhan serba boleh yang dianggap sebagai sumber yang boleh diperbaharui untuk serat semula jadi. Walau bagaimanapun, kanji termoplastik tulen juga mempunyai beberapa batasan, termasuk kekuatan mekanikal yang rendah, kestabilan jangka panjang, dan rintangan air, mengehadkan potensi aplikasinya. Sementara lilin sawit diiktiraf sebagai bahan hidrofobik yang sangat baik kerana ia berpotensi meningkatkan hidrofobik bahan. Untuk meningkatkan sifat kanji ubi kayu asli (CS), mengadun matriks kanji dengan bahan hidrofobik boleh meningkatkan prestasi biopolimer. Selain itu, gentian Cymbopogan citratus (CCF) merupakan pengukuhan yang berpotensi untuk komposit polimer. Oleh itu, pencirian gentian Cymbopogan citratus telah dijalankan untuk menganalisis potensinya sebagai bahan tetulang. Kemudian, beberapa kaedah pengubahsuaian telah digunakan untuk meningkatkan sifat kanji ubi kayu termoplastik (TPCS) iaitu; (1) pencampuran TPCS dengan lilin sawit (2) pengukuhan TPCS/PW dengan gentian serat Cymbopogan citratus, dan (3) rawatan alkali gentian serat CCF dalam adunan TPCS/PW. Justeru, komposit TPCS/PW/CCF telah berjaya dibangunkan dengan menggunakan kaedah mampatan acuan panas. Dari hasil keputusan diperolehi, menunjukkan bahawa sifat mekanikal bahan telah dipertingkatkan berikutan penggunaan lilin sawit. Sifat terma bahan telah bertambah baik sedikit apabila pemuatan lilin sawit meningkat daripada 0 hingga 15 wt%.Pembangunan dan pencirian TPCS dengan penggabungan 5 wt.% pemuatan lilin sawit menunjukkan kekuatan optimum sifat mekanikal. Selain itu, penambahbaikan dalam sifat mekanikal adunan TPCS/PW adalah bukti selepas penggabungan gentian serat Cymbopogan citratus. Keputusan menunjukkan bahawa sifat mekanikal gabungan TPCS/PW yang dipertingkatkan dengan penggabungan CCF, dengan kandungan CCF 50 wt.% menghasilkan modulus dan kekuatan maksimum. Ia juga terbukti daripada hasil analisis pembelauan sinar-X (XRD) bahawa indeks kehabluran komposit dipertingkatkan dengan penambahan CCF. Dari segi sifat terma, penambahan CCF meningkatkan kestabilan terma bahan, seperti yang ditunjukkan oleh suhu penguraian permulaan yang lebih tinggi dan kandungan abu. Selepas ditanam di dalam tanah selama 2 dan 4 minggu, penggabungan CCF ke dalam campuran TPCS/PW memperlahankan biodegradasi komposit. Akhir sekali, kesan rawatan alkali ke atas gentian Cymbopogan citratus ke dalam adunan kanji ubi kayu termoplastik/lilin sawit telah dinilai. Keputusan yang diperolehi menunjukkan bahawa, komposit yang dirawat menunjukkan peningkatan yang ketara dalam sifat mekanikal pada larutan NaOH 6% di mana sampel menunjukkan *masing-masing 19.9 MPa, 30.0 MPa, dan 13.3 MPa untuk kekuatan tegangan, lenturan dan hentaman. Ia juga terbukti bahawa, rawatan alkali gentian telah membawa kepada rintangan air yang lebih tinggi manakala hasil ditanam didalam tanah menunjukkan kebolehbiodegradasian yang lebih perlahan untuk komposit yang dirawat. Secara keseluruhan, dapatan daripada kajian ini menunjukkan bahawa TPCS yang diubah suai oleh lilin kelapa sawit, gentian Cymbopogan citratus, dan gentian dirawat alkali telah menunjukkan ciri fungsi yang lebih baik daripada bahan asal. Justeru, kajian ini mempertingkatkan potensi kanji ubi kayu termoplastik untuk dibangunkan sebagai alternatif bahan terbiodegradasi.*

ACKNOWLEDGEMENTS

Highest gratitude to Allah SWT for granting me the strength, resilience, and knowledge to complete this thesis. The successful completion of this thesis would not have materialised without His blessing and guidance. My deepest appreciation goes to my kind supervisor Dr. Ridhwan bin Jumaidin for the unending support and enthusiastic supervision he provided me throughout my PhD program. Not to forget, I would like to express my sincere appreciation for the guidance and coaching of my co-supervisors, Associate Professor Dr. Mohd Zulkefli Bin Selamat who always open his door whenever I ran into a trouble spot or needed advice with my writing or research.

I would like to convey my great appreciation to German-Malaysian Institute and Majlis Amanah Rakyat (MARA) for providing the scholarship award for me to pursue this study. Special appreciation to Universiti Teknikal Malaysia Melaka and the Ministry of Higher Education Malaysia for the research financial support through a research grant (RACER/2019/FTKMP-CARE/F00413).

I owe a special thanks to my dear husband Mohd Nazaril Issyam bin Ismail for his continuous encouragement and sacrifice throughout my study. To my beloved father (Kamaruddin bin Malan) and mother (Maimon binti Muhamad Nalan) for the motivations and continuous prayer for my success in this world and in the hereafter. Special credit to my sons (Muhammad Firaas Darwish and Muhammad Fareed Danish) for being such an understandable kids during my study period. Thanks to my siblings, Zatil Hazira, Zairul Hakimy, Zatil Hazrati, Zatil Hazika, Zatil Farisha and Zharif Hazrief for their constant moral support. It’s my fortune to cheerfully acknowledge the support of my entire family members including my in-laws (Ismail bin Ali Haniapiah and Siti Patona binti Ahmad).

Finally, I am thankful to all staff in the Faculty of Mechanical Engineering and Faculty of Mechanical and Manufacturing Engineering Technology for their help and assistance. Last but not least, my thanks go to all those people who knowingly or unknowingly helped me in the successful completion of this work such as Dr. Ahmad Ilyas bin Rushdan, Siti Nur Atikah binti Mahamud, sincere gratitude and thankful goes to my research colleague, Nur Diyana Binti Zakuan and Nurul Hanan Taharuddin for always being my supportive teammates, providing me guidance and knowledge sharing to successfully complete this work.

May Allah repay all your good deeds with kindness and ease in life. Thank you all.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

ASTM	-	American Society for Testing and Materials
ANOVA	-	Analysis of Variance
CH_3COOH	-	Acetic acid
CCF	-	*Cymbopogan citratus* fibre
CNC	-	Cellulose Nanocrystal
DSC	-	Differential Scanning Calorimetry
FTIR	-	Fourier Transform Infrared
FESEM	-	Field emission scanning electron microscope
HDPE	-	High-density polyethylene
KA	-	Alkali treated kenaf
KB	-	Bleached kenaf
LDPE	-	Low-density polyethylene
LGES	-	Lemongrass extracted solution
LGES/GO	-	Lemongrass extracted solution/ graphene oxide
LGDM/GO	-	Lemongrass dissolved mixture/graphene oxide
LLAC	-	Lemongrass leaves activated carbon
LLAC-MB	-	Lemongrass leaves activated carbon/methylene blue
MAPE	-	Maleic anhydride grafted polypropylene
MC	-	Moisture content
MA	-	Moisture absorption
NaOH	-	Sodium hydroxide

PW	-	Palm wax
PCL	-	Polycaprolactone
PE	-	Polyethylene
PP	-	Polypropylene
PHB	-	Polyhydroxybutyrate
PHBV	-	Polyhydroxybutyrate-co-valerate
PBS	-	Poly(butylene succinate)
PLA	-	Poly (lactic acid)
RH	-	Relative humidity
SEM	-	Scanning electron microscope
SPS	-	Sugar palm fibre
TGA	-	Thermal-gravimetric analysis
TPS	-	Thermoplastic starch
TPCS	-	Thermoplastic cassava starch
TS	-	Thickness swelling
W	-	Weight
WS	-	Water solubility
WVP	-	Water vapor permeability
XRD	-	X-ray diffraction