



UNIVERSITI PUTRA MALAYSIA

***MECHANICAL AND THERMAL CHARACTERIZATION OF
HYBRIDIZED SHORT KENAF/PINEAPPLE LEAF FIBER REINFORCED
HIGH DENSITY POLYETHYLENE COMPOSITES***

ISUWA SULEIMAN AJI

FK 2011 113

**MECHANICAL AND THERMAL CHARACTERIZATION OF
HYBRIDIZED SHORT KENAF/PINEAPPLE LEAF FIBER REINFORCED
HIGH DENSITY POLYETHYLENE COMPOSITES**



**Thesis submitted to the School of Graduate Studies, Universiti Putra
Malaysia, in fulfillment of the requirements for the Degree of Doctor of
Philosophy**

October 2011

DEDICATION

I would like to dedicate this work to my late father (Baba Stephen Aji Suleiman) and all other members of my immediate and extended family.

My wife (Dr Watirahyel Isuwa Aji), my two sons (Anjikwi-Barnabas Isuwa Aji and Musa-Gainako Isuwa Aji), to my mother who stood with me in prayers, my siblings Galadima and Mrs Dzarma Laushi, Mr and Mrs Maidoki, Mrs Saratu Bijimi, Mrs Hauwa Aji Ishaku, Mr and Mrs Suleiman Abba Aji, Mr and Mrs Isa Aji, my twin brother, (Yakubu Stephen Aji), my in-laws, Mr and Mrs Anjikwi Chiwar (OON), my family in Malaysia (PCC Equine Park), Bro. Vincent and Sister Amu Raj.

Thank you all for your patience, support and understanding.

Abstract of thesis submitted to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

MECHANICAL AND THERMAL CHARACTERIZATION OF HYBRIDIZED SHORT KENAF/PINEAPPLE LEAF FIBER REINFORCED HIGH DENSITY POLYETHYLENE COMPOSITES.

By

ISUWA SULEIMAN AJI

October 2011

Chairperson: Edi Syams Bin Zainudin, PhD

Faculty: Engineering

Hybrid composites of kenaf/pineapple leaf fiber (PALF) reinforced high-density polyethylene (HDPE) matrix were produced by compression molding operation. Tensile, impact and flexural tests as well as dynamic mechanical and thermogravimetric analyses (DMA and TGA) were performed to characterize the composites in variation to fiber loading, fiber length and kenaf/PALF fiber proportions. This is in addition to employing electron beam irradiation (EBI), use of compatibilizers/modifiers and crosslinkers in improving the composites mechanical properties. Characterisation of the composites was preceded with the optimization of the composite's processing parameters.

Hybridization has shown to provide better impact strength and reduction in overall water uptake of composites even without carrying out any chemical treatment. While PALF has improved the composite in tensile and flexural

properties, kenaf provided impact strength and reduction in the overall water uptake because of its better aspect ratio that ensured greater matrix dispersion in the composites. Furthermore, tensile and flexural properties of the hybrid are higher than that of the neat HDPE and this increase is in direct proportionality to increase in fiber loading for up to an optimum of 60% with 0.25mm fiber length; however, the impact strength of all un-modified hybrids was lower than that of neat HDPE except where EBI was employed at 10 kGy. Increasing fiber length did not show proportionate improvement in tensile and flexural properties, which could have been caused by fiber agglomeration, but impact strength showed otherwise.

Treatment of fiber surface with NaOH and Vinyltri(2-methoxy ethoxy) silane (silane AH172) and modification of HDPE matrix with the addition of MaPE and Poly (methylene) poly(phenil) isocyanate (PMPPIC) for the purpose of curtailing water uptake of the composite was successful. Irrespective of either fiber surface modification or matrix modification, reinforcement with respect to treatment depends on the type of modifier used and not the modification of matrix or fiber. Composites responded marginally to trimethylol propane trimethacrylate (TMPTMA) and silane that were employed as crosslinkers because HDPE self-crosslinked by radiation making silane and TMPTMA less effective, thus, radiating such composite without their addition is preferred. Thermal property from DMA results has shown that at lower temperatures, 60% fiber loading had reduced the loss modulus peak of the neat HDPE and

delayed the loss modulus of the hybrid up to about 100 °C. However, increasing the fiber content of the hybrid composite, raised the damping peak ($\tan \delta$) with increase in temperature. Thermogravimetric analysis (TG) and derivative thermogravimetric analysis (DTG) result showed that the main decomposition temperature occurred around 467 °C for all except composites prepared with 0.75 and 2 mm fiber length. There was a clear shift in decomposition temperatures of the composites with increase in fiber length while decomposition of hybrid composite is directly proportional to increase in fiber loading.

In conclusion, kenaf and PALF offered tremendous potential as hybrid fillers in HDPE matrix. They have shown to enhance thermal stability of composites, ease higher fiber loading vis-à-vis improved mechanical properties of matrix and reduction in water uptake even without treatment/compatibilization. This combination holds the edge for practical engineering application in automobile dashboard, side driving mirror casing and automobile door trim fabrication.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
Sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

**PENCIRIAN SIFAT MEKANIKAL DAN TERMA KOMPOSIT
POLIETILENA KETUMPATAN TINGGI DIPERTEGUH GENTIAN
HIBRID PENDEK KENAF / DAUN NANAS**

Oleh

Isuwa Suleiman Aji

October 2011

Pengerusi: Edi Syams Bin Zainudin, PhD

Fakulti: Kejuruteraan

Komposit polyetelin berketumpatan tinggi (HDPE) matriks diperteguh gentian hibrid kenaf-daun nanas (PALF) telah dihasilkan melalui operasi pengacuan mampatan. Ujian-ujian tegangan, hentaman dan lenturan serta analisis-analisis dinamik mekanikal (DMA) dan termogravimetri (TGA) telah dijalankan untuk pencirian hibrid dalam pelbagai kandungan gentian, panjang gentian dan nisbah gentian kenaf/PALF. Ini ialah sebagai tambahan kepada penggunaan Pancaran Sinar Elektron (EBI), penggunaan penyesuai/pengubahsuai dan penyilang dalam meningkatkan sifat-sifat mekanikal hibrid. Pencirian hibrid telah didahului dengan pengoptimuman parameter-parameter pemprosesan hibrid. Penghibridan telah terbukti memberikan kekuatan hentaman yang lebih baik dan pengurangan dalam kadar penyerapan air menyeluruh komposit walaupun tanpa melaksanakan mana-mana olahan kimia.

PALF membantu komposit dalam sifat-sifat ketegangan dan lenturan, manakala kenaf membekalkan kekuatan hentaman dan pengurangan yang menyeluruh dalam kadar penyerapan air disebabkan oleh nisbah aspeknya yang lebih baik dengan menjamin lebih penyerakan matriks dalam hibrid. Tambahan pula, sifat-sifat tegangan dan lenturan hibrid lebih tinggi daripada HDPE tanpa pengisi dan peningkatan ini berkadar langsung dengan peningkatan muatan gentian sehingga 60% muatan optimum dengan panjang serat 0.25 mm; bagaimanapun, kekuatan hentaman kesemua hybrid tanpa pengubahsuai berada lebih rendah daripada HDPE tanpa pengisi kecuali apabila pancaran sinar elektron telah dilakukan pada 10kGy. Penambahan panjang gentian tidak menunjukkan peningkatan yang berkadar langsung dengan sifat-sifat tegangan dan lenturan, ini mungkin disebabkan oleh pengumpalan gentian, walaubagaimanapun, kekuatan hentaman menunjukkan sebaliknya. Rawatan permukaan serat dengan NaOH and Vinyltri(2 metoksi etoksi) silana (silana AH172) dan pengubahsuaian matriks HDPE dengan penambahan MaPE dan Poly[metilena poly(fenol isosianat)] (PMPPIC) untuk tujuan mengurangkan kadar penyerapan air hibrid telah berjaya dicapai. Tanpa mengendahkan pengubahsuaian permukaan atau matriks, peneguhan bersandarkan rawatan bergantung kepada jenis pengubahsuai yang digunakan dan bukannya apa yang diubah. Hibrid bertindak balas secara marginal kepada TMPTMA dan Silana yang telah digunakan sebagai penyilang kerana HDPE dengan sendirinya telah tersilang disebabkan oleh pancaran sinaran yang menyebabkan Silana and TMPTMA

kurang berkesan, maka, pemancaran sinar ke atas komposit tanpa apa-apa penambahan adalah lebih diutamakan. Keputusan-keputusan sifat thermal dari (DMA) telah menunjukkan bahawa pada suhu-suhu yang lebih rendah, 60% kandungan serat telah mengurangkan puncak modulus kehilangan HDPE tulen dan melambatkan modulus kehilangan hibrid sehingga lebih kurang 100°C. Bagaimanapun, pertambahan kandungan gentian hibrid, menaikkan puncak redaman (tan delta) bersama dengan peningkatan suhu. Keputusan analisa thermogravimetric (TG) and analisa terbitan thermogravimetric (DTG) menunjukkan suhu penguraian utama berlaku adalah sekitar 467°C untuk semua hibrid kecuali hibrid yang menggunakan gentian yang panjangnya 0.75 dan 2 mm.

Terdapat satu anjakan yang jelas dalam suhu-suhu penguraian hibrid dengan peningkatan dalam panjang gentian manakala penguraian komposit hibrid adalah berkadar langsung dengan peningkatan dalam muatan gentian. Sebagai kesimpulan, Kenaf and PALF menawarkan potensi yang besar sebagai pengisi hibrid dalam matriks HDPE. Ia telah dibuktikan dapat meningkatkan kestabilan haba komposit, perbandingan yang setara bagi peningkatan kandungan gentian ialah meningkatnya sifat-sifat mekanikal dan pengurangan kadar penyerapan air walaupun tanpa rawatan/penyesuaian. Gabungan ini memberikan kelebihan bagi aplikasi kejuruteraan yang praktikal dalam pembuatan papan pemuka automobile, bingkai cermin pandang tepi dan fabrikasi perapi pintu automobil.

ACKNOWLEDGEMENT

Glory and honor to the creator of heavens and earth and everything therein.

By God's grace, I have been able to complete this research and the write up of my thesis in 4 semesters. I would like to express my deep appreciation to my supervisory committee Chairman, Dr Edi Syams bin Zainudin, for his unreserved support and encouragement that was unparalleled. I would also like to appreciate the support and understanding I got from my supervisory committee members, Professor Ir. Dr. Mohd. Sapuan Salit, Dr Khalina Abdan and Dr. Khairul Zaman Hj Moh'd Dahlan; thanks for being a pillar to my research. A big thanks to my lecturers, Professor Dr. Shamsuddin b. Sulaiman (Advance Manufacturing Technology), Assoc. Professor Parida Md Tahir (Lignocellulose Materials), Assoc. Professor Dr. Rashid b. Mohamed Shariff (Research Methodology), Assoc. Professor Dr. Zulkiflle b. Leman (Manufacturing Operation management), and Dr B.T Hang Tuah b. Baharudin (Manufacturing System Design). I will not forget to thank Mr Wan Ali of Nuclear Malaysia and Mr Muhammad Wildan Ilyas b. Mohamed Ghazali of Mechanical Engineering Laboratory, Universiti Putra Malaysia, for their unreserved assistance during my experiment in their Lab. Similar appreciation also to Dr Esther Gikonyo for academic support. To my friends Wan Hanifah, Firdaus Abdulrahman, Umar Abdul Hanan, Ridzwan Ishak, Mohd. Shukri, Sahari, Yousuf Ali El-Shekeil, Riza Wirawan and Dandi Bachtir, who have all made my stay in Malaysia interesting by supporting my study in UPM in one way or the other. My gratitude also goes to Tertiary Education Trust Fund (TETF) Nigeria for the financial support received.

APPROVAL PAGE

I certify that an Examination Committee met on **28th October 2011** to conduct the final examination of Isuwa Suleiman Aji on his thesis entitled "**Mechanical and Thermal Characterization of Hybridized Short Kenaf/Pineapple Leaf Fiber Reinforced High Density Polyethylene Composites**" in accordance with the Universities and University colleges act 1971 and the Constitution of Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the the Doctor of Philosophy.

Members of the Examination committee were as follows:

Professor Dr. Robiah bt Yunus

Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Professor Dr. Luqman Chuah Abdullah

Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Associate Professor Dr. Zulkiflle bin Leman

Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Dr. Michele Meo, PhD

Senior Lecturer
Faculty of Engineering and Design
Department of Material Design Center
University of Bath,
United Kingdom
(External Examiner)

.....
SEOW HENG FONG, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirements for the degree of Doctor of Philosophy. The members of the supervisory committee were as follows:

Edi Syams Bin Zainudin, PhD

Senior Lecturer

Faculty of Engineering

Universiti Putra Malaysia

(Chairman)

Mohd. Sapuan Salit, Ph.D, PEng

Professor

Faculty of Engineering

Universiti Putra Malaysia

(Internal Member)

Khalina Abdan, PhD

Senior Lecturer

Faculty of Engineering

Universiti Putra Malaysia

(Internal Member)

Khairul Zaman HJ M. Dahlan, PhD

Radiation Processing Technology

Malaysian Nuclear Agency

Bangi-Kajang

(External Member)

.....
BUJANG KIM HUAT, PhD

Professor and Dean

School of Graduate Studies

Universiti Putra Malaysia

Date:

DECLARATION

I declare that the thesis is my original work except for quotations and citations, which have been duly acknowledged. This thesis has also not been previously, and is not currently, submitted for any other degree at Universiti Putra Malaysia or other institution.

ISUWA SULEIMAN AJI
Date: 28 October 2011



TABLE OF CONTENT

| | Page |
|--|------|
| DEDICATION | ii |
| ABSTRACT | iii |
| ABSTRAK | |
| Error! Bookmark not defined. | |
| ACKNOWLEDGEMENT | ix |
| APPROVAL PAGE | x |
| DECLARATION | xii |
| LIST OF TABLES | |
| xviii | |
| LIST OF FIGURES | xix |
| LIST OF ABBREVIATIONS | |
| xxiv | |
| CHAPTER | 1 |
| 1 INTRODUCTION | 1 |
| 1.1 Overview | 1 |
| 1.2 Significance of the Study | 3 |
| 1.3 Problem Statements | 5 |
| 1.4 The Aim and Objectives of the Study | 8 |
| 1.5 Scope and Limitation of the Study | 8 |
| 1.6 The Outline of the Thesis | 9 |
| 2. LITERATURE REVIEW | 11 |
| 2.1 Polyethylene/HDPE | 11 |
| 2.2 Natural Fibers | 12 |
| 2.3 Kenaf | 18 |
| 2.3.1 Kenaf Composites | 19 |
| 2.4 Pineapple Leaf Fiber (PALF) | 21 |
| 2.4.1 PALF Composites | 23 |
| 2.5 Compatibility and Reinforcement Effect versus Surface Modification | 26 |
| 2.6 Hybridization | 32 |
| 2.7 Issues in Mechanical Properties of Composites | 34 |
| 2.8 Thermal Properties of Natural Fiber Composites | 35 |
| 2.9 Pertinent Reasons for Combining PALF and Kenaf Fibers | 36 |
| 2.10 Potential Application Areas | 38 |
| 2.11 Conclusion | 38 |
| 3. MATERIALS AND METHODS | 40 |
| 3.1 Materials | 40 |

| | |
|---|----|
| 3.2 High Density Polyethylene (HDPE) | 40 |
| 3.3 Machines and Equipment Used for Fabrication | 41 |
| 3.4 Mechanical Tests | 42 |
| 3.4.1 Tensile Testing of Composite | 42 |
| 3.4.2 Flexural Testing of Composite (Three-point bending) | 43 |
| 3.4.3 Impact Testing of Composite (Izod) | 45 |
| 3.4.3 Electron Beam Irradiation | 46 |
| 3.5 Production of Hybrid Composite for Mechanical Testing | 47 |
| 3.5.1 Fiber Treatment | 51 |
| 3.6 Water Absorption Test | 52 |
| 3.7 Testing and Test Specimen | 53 |
| 3.8 Thermal Property Test | 54 |
| 3.8.1 Dynamic Mechanical Analysis | 54 |
| 3.8.2 Fabrication and Testing of DMA Specimen | 56 |
| 3.8.3 Fabrication and Testing of TGA Specimen | 57 |
| 4. OPTIMIZING PROCESSING PARAMETERS FOR HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE | 59 |
| 4.1 Introduction | 60 |
| 4.2 Experimental | 62 |
| 4.3 Results and Discussion | 64 |
| 4.4 Summary | 69 |
| Copyright Permission Letter from KEM | 71 |
| 5. STUDYING THE EFFECT OF FIBER SIZE AND FIBER LOADING ON THE MECHANICAL PROPERTIES OF HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE | 72 |
| 5.1 Introduction | 73 |
| 5.2 Experimental | 74 |
| 5.2.1 Materials | 74 |
| 5.2.2 Preparation of Composite | 75 |
| 5.2.3 Mechanical Testing | 76 |
| 5.3 Results and Discussion | 76 |
| 5.3.1 Tensile Strength of Composite | 77 |
| 5.3.2 Flexural Result of Composites | 80 |
| 5.3.3 Impact Strength of Composite | 84 |
| 5.4 Conclusion | 86 |
| Copyright Permission Letter from JRP | 88 |

| | |
|---|-----|
| 6. ELECTRON BEAM IRRADIATION INDUCED CROSSLINKING OF HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE WITH AND WITHOUT CROSSLINKING AGENT | 89 |
| 6.0 Introduction | 90 |
| 6.2 Materials and Methods | 96 |
| 6.2.1 Materials | 96 |
| 6.2.2 Silane Fiber Treatment | 97 |
| 6.2.3 Preparation of Composite | 97 |
| 6.2.4 Electron Beam Irradiation (EBI) | 98 |
| 6.2.5 Mechanical Testing | 99 |
| 6.3 Results and Discussion | 99 |
| 6.4 Summary and Conclusion | 111 |
| Manuscript Acceptance Letter from JRPC | 113 |
| Copyright Permission Letter from JRPC | 114 |
| 7. STUDY OF HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE BY DYNAMIC MECHANICAL ANALYSIS | 115 |
| 7.1 Introduction | 116 |
| 7.2 Materials and Method | 119 |
| 7.2.1 Materials | 119 |
| 7.2.2 Preparation of Hybridized Composites | 120 |
| 7.3 Results and Discussion | 121 |
| 7.3.1 Effect of Varying Fiber-to-Fiber Ratios | 122 |
| 7.3.2 Effect of Fiber Loading on Damping Properties | 126 |
| 7.3.3. Effect of Fiber Length on Damping Properties | 129 |
| 7.4 Conclusion | 131 |
| Manuscript Acceptance Letter from PPTE | 133 |
| 8. THERMAL PROPERTY DETERMINATION OF HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE BY THERMOGRAVIMETRIC ANALYSIS | 134 |
| 8.1 Introduction | 135 |
| 8.2 Experimental | 137 |
| 8.2.1 Materials | 137 |
| 8.2.2 Preparation of Hybrid Composites | 138 |
| 8.2.3 Thermal Properties (Thermogravimetric Analysis, TGA) | 139 |
| 8.3 Results and Discussion | 139 |
| 8.3.1 Variations in Fiber Ratios | 139 |
| 8.3.2 Effect of Fiber Loading | 143 |
| 8.3.3 Effect of Fiber Length | 146 |
| 8.4 Conclusion | 148 |

| | |
|--|-----|
| Manuscript Acceptance Letter from JTAC | 150 |
| Copyright Permission Letter from JTAC | 151 |
| 9. ROLE OF FIBER/MATRIX MODIFICATION ON MECHANICAL PROPERTIES AND WATER SORPTION OF HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE | 152 |
| 9.0 Introduction | 153 |
| 9.2 Experimental | 159 |
| 9.2.1 Materials -Fibers and Matrix | 159 |
| 9.2.2 Coupling Agents | 159 |
| 9.2.3 Treatment | 160 |
| 9.2.4 Composite Preparation | 161 |
| 9.2.5 Water Absorption | 162 |
| 9.2.6 Mechanical Testing of Composites | 162 |
| 9.3 Results and Discussion | 163 |
| 9.3.2 Water Absorption | 173 |
| 9.4 Conclusion | 177 |
| 10. HYBRIDIZED KENAF/PALF REINFORCED HDPE COMPOSITE | 179 |
| 10.1 Introduction | 180 |
| 10.2 Materials and method | 185 |
| 10.2.1 Materials | 185 |
| 10.3 Method | 186 |
| 10.3.1 Preparation of Composite | 186 |
| 10.3.2 Mechanical Property Testing | 187 |
| 10.3.3 Water Absorption | 187 |
| 10.4 Results and Discussion | 188 |
| 10.5 Summary and Conclusion | 199 |
| 11. SUMMARY, GENERAL CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE RESEARCH. | 201 |
| 11.1 Summary | 201 |
| 11.2 General Conclusion | 213 |
| 11.3 Recommendations for Future Research | 214 |
| REFERENCES | 215 |
| APPENDIX A Proof that Key Eng'g Materials is indexed in Scopus | 236 |
| APPENDIX B Proof that Journal of reinforced plastic and composite is indexed in ISI Science citation Index Expanded | 237 |
| APPENDIX C Proof that Journal of Thermal Analysis and Calilometry is indexed in ISI Science citation Index Expanded | 238 |

| | |
|--|------------|
| APPENDIX D Proof that Journal of Polymer-Plastic Technology and Engineering is indexed in ISI Science citation Index Expanded | 239 |
| APPENDIX E Proof that Journal of Composite Materials is indexed in ISI Science citation Index Expanded | 240 |
| APPENDIX F1 Chapter 10 SAS System- GLM Procedure | 241 |
| APPENDIX F2 Chapter 5 SAS System- GLM Procedure | 243 |
| APPENDIX F3 Chapter 6 SAS System - GLM Procedure | 246 |
| BIODATA OF STUDENT | 248 |
| LIST OF PUBLICATIONS | 249 |

