INVESTIGATION ON FLAX/KEVLAR HYBRID WITH POLYLACTIC ACID LAMINATED COMPOSITES BY INCLUSION OF NANOCLAY AND GRAPHENE

By

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Chair : Norkhairunnisa binti Mazlan, PhD
Faculty : Engineering

Due to the low mechanical properties of the natural fiber, hybridisation of flax and kevlar may benefits the use of only single fiber in improving the mechanical properties of the composite materials. Furthermore, inclusion of sufficient amount of nanofillers such as graphene or nanoclay in a fiber reinforced polymer composite can aid to improve the flexural and impact properties of the composite. In this research, hybrid flax/kevlar fibre composite reinforced with PLA composites have been prepared by using hot press technique. Initially, the flax fibre were treated with 2 % of sodium hydroxide (NaOH) solution. Nanoclay and graphene were sonicated separately at designated concentration with chloroform and then stirred together with PLA pellet to produce a thin film. The main objectives of this research is to investigate the properties hybrid fibre laminated composites with addition of nanofillers. The specific objectives focused on the investigation of flexural properties of the hybrid laminated nanocomposites with presence of different nanofillers loading. Next, investigate different impact energy of low velocity impact response on the hybrid laminated nanocomposites and finally to analyse the failure morphology of hybrid laminated nanocomposites. Flexural test revealed improvement in strength. Flax/kevlar/PLA nanocomposite filled with 1 wt.% of nanofiller shows improvement in flexural strength of about 13.38 and 8.67 %, for nanoclay and graphene compared to composite without nanofillers. From scanning electron microscope (SEM) images, rough fractured surface was identified from the flexural failure, which due to the slow spreading crack and high energy absorption of the flax/kevlar/PLA composite to resist failure. Impact tests at 10 and 20 J of impact energy for the flax/kevlar/PLA nanocomposite shows incomplete punching holes at the bottom surface layers while complete penetration was found for the flax/kevlar/PLA nanocomposite during impact

energy level at 30J. Inclusively, addition of nanofillers improve the mechanical properties up to 1 wt.% while increasing it over 3 wt.% will decrease the properties of composites.

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

PENYIASATAN TERHADAP HIBRID *FLAX/KEVLAR* DENGAN KOMPOSIT ASID POLIKLASTIK BERLAMINA DENGAN PENYERTAAN *NANOCLAY* DAN *GRAFENE*

Oleh

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Disebabkan oleh sifat mekanikal serat semula jadi yang rendah, penghibridan *flax* dan *kevlar* boleh memanfaatkan penggunaan serat tunggal sahaja dalam meningkatkan sifat mekanikal bahan komposit. Tambahan pula, kemasukan jumlah pengisi *nano* yang mencukupi seperti *graphene* atau *nanoclay* dalam komposit polimer diperkuat serat boleh membantu meningkatkan sifat lentur dan kesan komposit. Dalam penyelidikan ini, komposit gentian *flax/kevlar* hibrid yang diperkukuh dengan komposit *PLA* telah disediakan dengan menggunakan teknik penekan panas. Pada mulanya, gentian rami telah dirawat dengan 2% larutan natrium hidroksida *(NaOH). Nanoclay* dan *graphene* diolah secara berasingan pada kepekatan yang ditetapkan dengan kloroform dan kemudian dikacau bersama pelet *PLA* untuk menghasilkan filem nipis. Objektif utama penyelidikan ini adalah untuk menyiasat sifat komposit berlamina serat hibrid dengan penambahan pengisi *nano*. Objektif khusus tertumpu pada penyiasatan sifat lenturan nanokomposit berlamina hibrid dengan kehadiran pemuatan pengisi *nano* yang berbeza. Seterusnya, siasat tenaga impak berbeza bagi tindak balas impak halaju rendah pada nanokomposit berlamina hibrid dan akhirnya untuk menganalisis morfologi kegagalan nanokomposit berlamina hibrid. Ujian lentur menunjukkan peningkatan dalam kekuatan. Nanokomposit flaks/kevlar/PLA yang diisi dengan 1 wt.% pengisi nano menunjukkan peningkatan dalam kekuatan lentur kira-kira 13.38 dan 8.67 %, untuk *nanoclay* dan *graphene* berbanding komposit tanpa pengisi *nano*. Daripada pengimbasan imej mikroskop *elektron (SEM*), permukaan patah kasar dikenal pasti daripada kegagalan lenturan, yang disebabkan oleh rekahan merebak yang perlahan dan penyerapan tenaga tinggi komposit *flax/kevlar/PLA* untuk menahan kegagalan. Ujian impak pada 10 dan 20 J tenaga hentaman untuk nanokomposit *flax/kevlar/PLA* menunjukkan lubang tebukan yang tidak lengkap pada lapisan

permukaan bawah manakala penembusan lengkap ditemui untuk nanokomposit *flax/kevlar/PLA* semasa tahap tenaga hentaman pada 30J. Secara inklusif, penambahan pengisi *nano* meningkatkan sifat mekanikal sehingga 1 wt.% manakala meningkatkannya melebihi 3 wt.% akan mengurangkan sifat komposit.

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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:

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LIST OF ABBREVIATIONS

-CH3	Methyl side group
2D	2 dimensional
CNSL	Cashew nut liquids
CO2	Carbon dioxide
EFB	Empty fruit bunch
FF	Flax fibre
G	Graphene
GPa	Gigapascal
HDPE	High density polyethylene
KE	Kevlar epoxy
KOH	Potassium hydroxide
LiOH	Lithium hydroxide
MPa	Megapascal
MWCNT	Multi walled carbon nanotube
NaOH	Sodium hydroxide
NC	Nanoclay
PE	Polyethylene
PHB	Polyhydroxybutyrate
PLA	Polylactic acid

CHAPTER 1

INTRODUCTION

1.1 Background of study

Inappropriate waste disposal and treatment is one of the most severe environmental challenges most countries face. The increasing globalisation and modernisation will likely further increase waste generation for disposal. Biocomposite products discarded by the user during use will become biocomposite wastes shortly, and these products will naturally degrade in the environment due to air, moisture, climate, or soil. However, when the number of biodegradable materials in the environment grows, the biodegradability of biocomposites can be improved by applying additives during the production process and concentrating on the products' end-uses. Importantly, proper waste disposal systems do not exist in many parts of the world, and wastes are simply dumped in the surrounding areas.

PLA is the most promising and widely used material with the best growth prospects, and it is also considered as a "green" environmentally friendly material. PLA possesses exceptional qualities including high stiffness and strength. It is used in many applications, including consumer packaging, biodegradable plastic bags, and automotive applications (Jonoobi et al., 2010). As a result of growing environmental awareness, fabrication and the used of traditional composites comprised of synthetic fibers such as aramid, glass, or carbon fibre as reinforcing materials have been challenged recently. Green composites are named from the fact that they are made of natural fibres and biopolymers that are entirely biodegradable and offer environmental advantages (Mohanty et al., 2005).

Recent developments in natural polymer composites have heightened the need to improve its mechanical properties and one of the solutions is the utilization of woven laminated fibre as reinforcement. Laminated woven fibre promotes higher fibre packing and offers good toughness through its mechanical interlocking (Alavudeen et al., 2011). The laminated woven fibre are consistent structures that combine the composition and characteristics of two or more compatible materials physically way and in some cases in shape. While Kelly (1967) emphasizes that, it is not a simple combination of two materials that the composites should be considered. The mixture has its unique characteristics in the wider context.

Biocomposite reinforcement refers to natural fibres such as flax, jute, kenaf, hemp, pineapple, sisal, cotton, banana, ramie, etc. Natural fibres are known for their quality, renewability, low density, low cost, and acceptable fibre properties, making them an appealing, sustainable alternative to the

traditional synthetic fibres used to manufacture composites. Natural composite fiber is used in transport (automobiles, trains, aerospace), military applications, consumer products, building industries and construction (ceiling panels, partition boards), packaging, etc. and more environmentally friendly applications.

1.2 Problem statement

Plastics are used in several industries today, including packaging, general manufacturing, pharmaceutical and medical manufacturing. However, because of they pollute natural resources and take a long time to degrade, the widespread usage of conventional petrochemical plastics pollutes the atmosphere. When it comes to this issue, the emphasis is on using materials that degrade naturally in the environment. PLA has been an attention in this reseach because it is currently the most important synthetic biodegradable polymer in spite of its mechanical property limitations. The addition of nanofiller to PLA is suggested to improve the polymer's mechanical properties, such as strength, mechanical modulus, and toughness. Nanoclay has high thermal stability and chemical resistance while graphene can produce a drastic amendment in the properties at very low filler content. In addition, laminated fibre will create a balance between environmental impact and performance with addition of synthetic and natural fibre. Despite their commonness, very few composites are fabricated using the solvent casting process. Therefore, this study was conducted on the solvent casting technique to understand PLA mechanical properties further. There also many ways to increase the properties of PLA. One of the methods is to use a laminated system. However, there is lack of information on the properties of hybrid flax/kevlar fibre laminated composites. No information on flax/kevlar hybrid with PLA laminated composites with addition of nanofillers motivates this study to be conducted. This study was design to focus on effect of laminated fibre composites with addition of nanofillers on its mechanical strength.

1.3 Research objectives

This research aims to analyse the different loading of different types of nanofillers reinforced with flax/kevlar hybrid in PLA. Therefore, the specific objectives of this research were as follows:

1) To study flexural properties the flax/kevlar hybrid PLA laminated nanocomposites with different nanofillers loading.
2) To investigate different impact energy of low-velocity impact response on the flax/kevlar hybrid PLA laminated nanocomposites.
3) To analyse the failure morphology of flax/Kevlar hybrid PLA laminated nanocomposites.

1.4 Scope and Limitation of the Study

The scope of this research focuses on the lamination of flax/kevlar hybrid PLA with the inclusion of nanofillers. Composites are produced with nanoclay and graphene to compare the effect of the weight percentage of nanofillers in composites. Parameters for optimum nanoclay and graphene were based on the preliminary study`s of best flexural and impact strength properties. The effect of nanoclay and graphene in PLA laminated composites was studied with loadings of 1.0 wt%, 3.0 wt% and 5.0 wt%. The stacking sequence of flax and kevlar fibre is limited to Kevlar/Flax/Kevlar/Flax/Kevlar (KFKFK). Thus all mechanical properties analysed are based only on one type of fibre arrangement. The mechanical properties for all of the composites produced were studied via flexural and low velocity impact tests. Morphology and chemical bonding of the samples were investigated using a scanning electron microscope (SEM), and fourier transform infrared (FTIR), respectively which are used to support the mechanical test result.

1.5 Thesis layout

This thesis consists of five chapters where all the content is summarised at the end of each chapter. The first chapter introduces the brief background of the composite materials application, the problem statement, the aim of this study, research objectives, the scope of the study and the thesis layout. The second chapter reviews and classifies the fibre used in this research. Its presents a literature review of composite material manufacturing and its effects on the mechanical behaviour of composite products. The failure modes in the composite were reviewed as well in this chapter. Chapter 3 describes the materials and methodology used in this study. It focus on a specific approach in designing and planning the course of study, and several experimental test procedures for this research. Materials characterisation conducted in this work, such as flexural and low-velocity impact, were explained.

Chapter 4 presents the results of physical and mechanical properties of the flax fibre with and without alkaline treatment, plain woven flax/Kevlar, flax/Kevlar hybrid with graphene and flax/Kevlar hybrid with nanoclay. In addition, the effect of different loading of graphene and nanoclay on the drop weight properties was discussed. Furthermore, the morphological properties of the fractured surface of the hybrid nanocomposites were also analysed. Furthermore, this chapter encompasses the ballistic impact properties results of the hybrid nanocomposites materials. Finally, Chapter 5 summarises the overall findings obtained from current study and recommendations for future research studies.

1.6 Significant of study

In this present study, the research contributions were as follows:

a) An exhaustive literature review found no information on the procedure for fabrication of PLA thin film using the solvent casting method. Thus, this study provides the solvent casting method to fill the gap.
b) There was a lack of information in a thorough review of the literature to fabricate the flax/kevlar hybrid in PLA film with nanocomposites using the hot press technique. As a result, this research proposes a solvent casting approach to bridge the gap.
c) Lack of study on comparison of different loading with a different type of nanocomposites in terms of impact load, impact energy and penetration resistance applications in the composites have been made. Therefore, this study focuses on the effect of the different types of nanocomposites adding to the composites.

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