MECHANICAL AND WATER ABSORPTION PROPERTIES OF HYBRID KENAF AND PINEAPPLE COMPOSITE ADDED WITH EPOXIDIZED NATURAL RUBBER

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Thank you God for blessing me with the presence of loving person around me.

To my husband… your love and your support along this journey have helped me faced the challenges that spread ahead,

To my children… thank you for standing by Umi and be very understanding,

This dedication also goes to my sisters, brothers, my mother…
… and my late father who always live in my memory… you brought me up and made me the person I am today.

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ABSTRACT

Wood polymer composites, WPC, is a competitive material which ranging from consumer products to engineering parts in various of application field. In this research, mechanical properties and water absorption were investigated on the hybrid WPC made from kenaf fibre and pineapple leaf fibres, PALF, as fillers at three level of total fibre loading of 30 %, 40 % and 50 % by weight, mixed in high density polyethylene, HDPE. The fibres hybrid ratio was kept constant at 60% kenaf to 40% PALF and the composite was compounded in a melt mixer and fabricated by compression moulding. The effect of different total fibres loading and addition 3% by weight epoxidized natural rubber, ENR, into the composite formulation was evaluated. For both with and without addition of ENR into kenaf-PALF/HDPE composite, tensile strength, tensile modulus, flexural modulus, impact strength and water absorption increased with an increase in total fibres loading, but the elongation at break of the composite decreased with increasing total fibres loading. Flexural strength only increased at lower fibre loading but decreased at highest fibre loading of this experiment. Meanwhile, overall effect of ENR addition was that it enhanced the tensile strength, impact strength and water absorption of the composite but only improved flexural strength and flexural modulus at lower fibre loadings of 30 and 40 %. On the other hand, ENR decreased the tensile modulus and elongation at break of the composites. At highest fibres loading from this study and with 3 % ENR-50 added, the composite, KP50PE2 is deformable and experienced a slight decreased in tensile strength. However, its enhanced impact property and higher toughness enabled the composite to withstand impact loading.

ABSTRAK

Komposit Polimer Kayu, WPC, adalah bahan yang kompetetif yang digunakan di dalam produk pegguna sehinggalah ke barangan kejuruteraan di dalam pelbagai lapangan applikasi. Di dalam kajian ini, sifat-sifat mekanikal dan penyerapan air adalah dikaji ke atas hibrid WPC yang dibina dari hibrid di antara gentian kenaf dan gentian daun nanas, PALF yang berperanan sebagai pengisi pada tiga paras jumlah pengisian gentian iaitu 30 %, 40 % and 50 % mengikut berat, yang dicampur dengan polietilena berketumpatan tinggi, HDPE. Nisbah hibrid gentian adalah ditetapkan pada 60% kenaf kepada 40% PALF dan komposit dibancuh di dalam pencampur dan difabrikasi menggunakan pengacuan mampatan. Kesan jumlah pengisian gentian yang berbeza dan penambahann 3% mengikut berat getah asli terepoksidakan, ENR, ke dalam formulasi komposit dinilai. Bagi kedua-dua formulasi dengan dan tanpa penambahan ENR ke dalam komposit kenaf-PALF/ HDPE, kekuatan regangan, modulus regangan, modulus lenturan, kekuatan hentaman dan penyerapan air bertambah dengan penambahan di dalam jumlah pengisian gentian, tetapi peratus pemanjangan komposit ketika putus berkurang dengan penambahan di dalam jumlah pengisian gentian. Kekuatan lenturan hanya bertambah pada jumlah pengisian gentian yang rendah tetapi berkurang pada pengisian gentian yang tinggi. Manakala kesan keseluruhan apabila ENR ditambahkan, adalah ia telah meningkatkan kekuatan regangan, kekutan hentaman dan penyerapan air oleh komposit tetapi hanya memperbaiki kekuatan lenturan dan modulus lenturan pada pengisian gentian yang rendah iaitu pada 30 and 40 % pengisian gentian. Sebaliknya, ENR mengurangkan modulus regangan komposit dan mengurangkan peratus pemanjangan komposit ketika putus. Pada pengisian gentian tertinggi dari kajian ini dan dengan penambahan 3 % ENR-50, komposit dengan formula KP50PE2 menunjukkan ciri mudah berubah dan hanya melalui perubahan yang sedikit sahaja dalam kekuatan regangan. Walau bagaimanapun peningkatan dalam ciri hentamannya dan pertambahan ketahanan membolehkan komposit tersebut menghadapi kekuatan hentaman.

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LIST OF ABBREVIATIONS

ASN	-	Atlas Automatic Sample Notcher
ASTM	-	American Society for Testing and Materials
BS	-	British standard
$CaCO_3$	-	Calcium carbonate
DCP	-	Dicumyl peroxide
E-GMA	-	Ethylene-glycidil methacrylate
EN	-	European standard
ENR	-	Epoxidized natural rubber
ENR-25	-	Epoxidized natural rubber at 25 % mol of epoxidation level
ENR-50	-	Epoxidized natural rubber at 50 % mol of epoxidation level
FTIR	-	Fourier-transform infrared spectroscopy
HDPE	-	High density of polyethylene
LENR	-	Liquid epoxidized natural rubber
LNR	-	Liquid natural rubber
MAPE	-	Maleated polyethylene
MAPP	-	Maleic anhydride-grafted polypropylene
NaOH	-	Sodium hydroxide
NBR	-	Nitrile rubber
NR	-	Natural ruber
NFM	-	Non-fibrous material
OH	-	Hydroxyl group
PALF	-	Pineaapple leaf fibre
PA6	-	Nylon 6
PBAT	-	Poly(butylene adipate-co-terephthalate)

PE	-	Polyethylene
PE-g-MA	-	Maleated polyethylene
PET	-	Polyethylene terephthalate
PHBV	-	Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate)
PLA	-	Polylactic acid
PP	-	Polypropylene
PVC	-	Polyvinyl chloride
RNP	-	Recycled newspaper
SEM	-	Scanning electron microscopy
Si	-	Silica
TPU	-	Thermoplastic polyurethane
UHMWPE	-	Ultra high molecular weight of polyethylene
UPR	-	Unsaturated polyester resin
WGL	-	Whole ground pineapple leaf
WPC	-	Wood polymer composite

LIST OF SYMBOLS

ρ_c	-	Density of composite
ρ_{HDPE}	-	Density of HDPE
ρ_{kenaf}	-	Density of kenaf
ρ_{PALF}	-	Density of PALF
g/10 min	-	grams in 10 minutes
g/cm^3 (gcm^{-3})	-	grams per centimeter cubic
kJ/m^2	-	Kilojoule per meter square
kN	-	KiloNewton
MPa	-	Megapascal
m_c	-	Mass of composite
m_{HDPE}	-	Mass of HDPE
m_{kenaf}	-	Mass of kenaf
m_{PALF}	-	Mass of PALF
mm^3	-	Milimeter cubic
mm/min	-	Milimeter per minute
Nm	-	Newton meter
T_g	-	Glass transition temperature
V_c	-	Volume of composite
V_{HDPE}	-	Volume of HDPE
V_{kenaf}	-	Volume of kenaf
V_{PALF}	-	Volume of PALF
w_1	-	Initial weight at day 1
w_2	-	Final weight after 24 hours immersion

w_{uptake}	-	Weight gained by the composite
w/v	-	Weight per volume

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CHAPTER 1

INTRODUCTION

1.1 Road safety barriers

Guardrails are road barriers of a safety devices fixed at the roadside or at highway (Xiao and Shang, 2011; Tabatebaei et al., 2012), that is meant to ensure the safety on the public roads (Borovinsek, 2013).

"Guardrail is longitudinally placed elements which are connected by bolts to guardrail connector where guardrail connector is an element for the connection of the neighbouring guardrails using bolt connection. Guardrail are deformable and reduce the severity of an impact but should be strong enough not to rupture during vehicle impact" (Borovinsek, 2013).

Among the material that are suitable for use in road safety application are plastics, fibreglass, rubber, wood fibres and they were used either alone or in various combination (Bligh et. al., 1995). In this study, wood polymer composite,WPC synthesised by using hybridised fibre based on pineapple leaf fibre (PALF) and kenaf fibre in high density of polyethylene (HDPE) matrix was suggested as an alternative material for guardrail and the material is expected to be able to experience deformation easily but sufficiently tough to withstand an impact.

1.2 Wood polymer composite

Wood polymer composite (WPC) is gaining much attention from the industry and becomes a dynamic sector in plastic industry (Amir and Ashori, 2010). The trend in WPC's product is developing product range that is having an enhancement in terms of its mechanical properties and durability. "*Major growth in the technology is coming from equipment design, process formulation and durability"* (Niska and Sain, 2008).

WPC has two main constituents, polymer and wood (Amir and Ashori, 2010). Wood are usually in the form of chips, flour or special wood fibres; whereas the plastics usually consumed are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene as a matrix in a virgin or recycled formed (Carus and Gahle, 2008; Cai et al., 2012). "*WPC are thermoplastically processible composite that consist of varying contents of woods, plastics and additives, and are processed by thermoplastic shape-forming techniques such as extrusion, injection moulding, rotomoulding or pressing"* (Carus and Gahle, 2008).

The natural fibre from wood as composite material in WPC have advantages in terms of its specific properties such as high specific strength and modulus, low density property, non-abrasiveness or low friction during processing (Amir and

Ashori, 2010; Kim and Pal, 2010), and in addition, they are also a low-cost material for replacing synthetic fibres, non-toxic, renewable and are environmental friendly (Amir and Ashori, 2010; Perez-Fonseca, 2014; Gouda, 2014). Natural fibres are used in many fields, for example, a natural fibre reinforced composite for automotive application such as door panels, seat backs, armrests, consoles and other part shifting form using steel and aluminium material (Zampaloni, 2007; Akil, 2015), sisal fibre reinforcement epoxy resin composite for orthopaedic implants (Gouda, 2014),

1.3 Hybrid wood polymer composite

When two or more different types of reinforcement applied in a matrix, hybrid composites is produced. Hybrid composites composed with two different fibres nowadays is harbouring interest scientifically and industrially (Beauson, 2013). Hybrid composites are developed for synergism effect that are able to display by the chosen fillers and matrix (Ashori and Sheshmani, 2010). Each fibres used must have different mechanical properties and different interaction with matrix, thereby some advantages of properties can be gained from both fibres utilized (Perez-Fonseca et al., 2014; Beauson et al., 2013; McDonough et al.,2004). Therefore, with appropriate combination of two natural fibres will result in properties improvement of composite material and at the same time maintain the environment natural appeal (Perez-Fonseca et al., 2014; Beauson et al., 2013).

1.4 Problem statement

Safety barriers or guardrails is a method for roadside immunization where the deviated vehicles will be redirect onto right path, preventing overturning that might cause collision with dangerous obstacle along the road sides, hence, reduce fatality and injury. However, there were also studies that showed that most death accidents involving collision with safety barriers especially guardrails are due to technical and structural faults, improper instalment place and improper design (Tabatebaei et al., 2012). Such casualty and fatal could be avoided if some investment and effort is made in providing new design in terms of material used in constructing a safer guardrail. Diagrams showing a fatal accident between vehicles and guardrail where guardrail had penetrated into the vehicles and cause injuries to the passenger are shown in Appendix A.

According to the British standard BS EN 1317-2:1998 and also a study reported by Atahan et al., 2014 on road restraint system, safety barrier should behave as the following:

> *"The safety barrier shall contain and redirect the vehicle without complete breakage of the principal longitudinal elements of the system.*
> *No major part of the safety barrier shall become totally detached or present an undue hazard to other traffic, pedestrians or personnel in a work zone.*
> ***Elements of the safety barrier shall not penetrate the passenger compartment of the vehicle.***
> ***Deformations of, or intrusion into the passenger compartment that can cause serious injuries are not permitted.***

Ground anchorages and fixings shall perform according to the design of the safety barrier system".

Interaction between drivers, vehicles, road and environment determines the highway design safety, and the comfort operating of the highway, where ensuring these safety and comfort for the user has become a growing concern (Wei, 2012). In 1995, Bligh et. al. had reported that products design to serve as direct substitute to currently used wood and steel in guardrails are lacking. Nowadays, some study had been conducted in finding new material of roadside barrier system which in present is using conventional metal, concrete or based on steel w-beam guardrail in order to withstand without failure the load coming from the impact of the crashing vehicles (Xiao and Shang, 2011; Viderkers et al., 2013; Borovinsek, 2013).

If car manufacturers are nailing interest in part components made from natural fibres composite (El Messiry, 2013), the road safety devices could also take the same action in contributing to safer and healthier environment by considering an alternative material for guardrail. Concrete barrier of road safety devices for instance, leaves more damages for being very rigid (Tabatebaei et al., 2012).

Wood polymer composite made from hybridized fibres of kenaf-pineapple leaf fibres in high density polyethylene, were considered in this study for alternative material of guardrail or road barrier system. The kenaf fibre has the ability to toughen and improve impact property of the guardrail due to its high aspect ratio (Aji et al., 2012; Akil et al., 2015) that gives an advantage in spreading the matrix throughout the composite, therefore yields in a better stress transfer from the matrix to fibre (Aji et al., 2012). On the other hand, high tensile strength of PALF will increase flexibility where it is achieved because PALF's cellulose has lower microfibrillar angle with ribbon like orientation that provides spring-like extension in tension and compression (Aji et al., 2012). In addition, kenaf fibre reduced water absorbance in the hybrid composite since it has lower cellulose content (Aji et al., 2012; Akil et al., 2015).

Appropriate blending and correct intermingling of bast (stem) of kenaf and leaf fibre of PALF are expected to give an optimum performance that has stiffness-toughness balance meant for composite to be used for structural application. Aji et al, (2012) reported that at equal ratio of kenaf and PALF, hybridisation effect had optimized tensile and flexural properties of the composite with HDPE matrix. At 60 % kenaf loading, impact strength was optimised. The hybridized fibres are able to give synergistic relationship in the composite without their properties being deteriorated when combined with the matrix, and at certain hybrid formulation between kenaf and PALF, interfacial bonding of fibres to the matrix was improved, thus yielded a hybrid composite with effective mechanical properties.

In this study, the composite is using thermoplastic resin of HDPE which allows the potential of being remoulding and recycle. Epoxidized natural rubber at 50 % epoxidation level, ENR-50 was added to the composite formulation to act as additional absorbent of energy and prolong the elastic deformation, thus is expected to contribute in improving shock-absorbent capacity. The fabricated composites were then be investigated for its deformable property and toughening effect.

1.5 Objective of the study

1. To investigate the mechanical properties and water absorption of kenaf/ PALF fibres filled HDPE composite at different fibres loading.

2. To evaluate the mechanical properties and water absorption of the composite after mixing with 3 % ENR-50.

1.6 Scope of the study

The kenaf fibres and PALF were randomly oriented, mixed, and used as fillers in high density polyethylene. Toughening agent of epoxidized natural rubber at 50 % mol of epoxidation level (ENR-50) was incorporated in the composite. The hybridized formula of kenaf and PALF as fillers, was kept constant at ratio 60 % kenaf: 40 % PALF and the WPC was formulated at specific fibre loading. HDPE is the matrix in this preparation of WPC.

The main parameters investigated was how different percentage weight of filler content in mixing ratio of fibrous composite formulation; and the effect of addition 3 % by weight ENR-50 in the composite formula will affect the properties of the hybrid WPC produced. The mechanical properties studied are tensile, flexural and impact properties. Composite's water uptake behaviour after 24 hours immersion in water for 11 days were also investigated. ENR ability in enhancing WPC's mechanical properties and its ability to resist water uptake were evaluated.

1.7 Significance of study

The development of incorporating lignocellulosic material in thermoplastic matrix are favourable (Talavera et al., 2007) and are giving a great impact on environmental and social issues (Monteiro, 2011). The material has high strength to weight ratio property, low deformation character, highly energy-absorbent and is cost saving (Xiao and Shang, 2011; El Messiry, 2013). The composite with lignocellulosic material in its formula is renewable, thus supporting an effort of recycling an agriculture waste (El Messiry, 2013; Talavera et al., 2007). In addition, the ability of the recyclability of plastic lead to positive eco-environmental impact (Talavera et al., 2007).

Mechanical properties of fibre reinforced composite materials can be tailored to serve the application outlined for the end products by changing the orientation or placement of the fibre. (Zampaloni, 2007). Therefore, kenaf-PALF in HDPE WPC is offering new materials for guardrails, where HDPE consumption is offering a cost reduction compared to the use of ultra high molecular weight polyethylene, UHMWPE, suggested by Xiao J. and Shang X. (2011) for guardrail material's substitution. The materials designed for road safety devices should be able to move or deviate at "working width" during the impact. In this study, ENR-50 was added into the composite for toughening reason. The capacity to be able to absorb impact energy through deformation of the guardrails will reduce the severity of the crash of collision (Tabatebaei et al., 2012). Hence, by adding ENR, the complete interlayer interfacial of ENR towards the fibres is expected to improve fracture toughness of the composites (Tanjung et al., 2015).

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