Research Article

Hybrid Fibre Polylactide Acid Composite with Empty Fruit Bunch: Chopped Glass Strands

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Received 26 June 2014; Revised 27 September 2014; Accepted 29 September 2014; Published 14 October 2014

Academic Editor: Hui Shen Shen

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Hybrid polylactide acid (PLA) composites reinforced with palm empty fruit bunch (EFB) and chopped strand E-glass (GLS) fibres were investigated. The hybrid fibres PLA composite was prepared through solution casting followed by pelletisation and subsequent hot compression press into 1 mm thick specimen. Chloroform and dichloromethane were used as solvent and their effectiveness in dissolving PLA was reported. The overall fibre loading was kept constant at volume fraction, V_f , of 20% while the ratio of EFB to GLS fibre was varied between V_f of 0 : 20 to 20 : 0. The inclusion of GLS fibres improved the tensile and flexural performance of the hybrid composites, but increasing the glass fibre length from 3 to 6 mm has a negative effect on the mechanical properties of the hybrid composites. Moreover, the composites that were prepared using chloroform showed superior tensile and flexural properties compared to those prepared with dichloromethane.

1. Introduction

Fibre reinforced composites based on carbon, glass, and Kevlar have been widely used in the aviation, automotive, marine, sport, and defence industries, attributed to their high strength to weight ratio, easy formability, and high tensile and fracture resistance. However, synthetic fibres are generally manufactured through energy intensive processes that produce toxic by-products while their reinforced composites are difficult to recycle and resistant to biodegradation [1]. Increasing governmental pressure as well as consumer and industrial awareness on the long-term effect of environmental pollution due to noncompostable polymeric products has led numerous researchers around the world to have gained interest to develop greener composites by either eliminating or minimising the usage of nondegradable synthetic polymeric resin and fibres.

Biodegradable polymeric resins generally can be categorised into two groups depending on their origin, natural biopolymers (polymer derived from natural resources such as starch, cellulose, gelatine, casein, wheat gluten, silk, wool,

plant oils, and polylactic acid), and synthetic biopolymers (mineral based biopolymer synthesised from crude oil with example including aliphatic polycaprolactone, aromatic polybutylene succinate terephthalate, and polyvinyl alcohols). Amongst the many natural-origin biodegradable polymers, polylactic acid (PLA), a corn-based biodegradable polyester obtained from fermentation of sugar feedstock, is gaining its popularity in the scientific community [2–4] and was chosen as the binding matrix material in this work.

Motivated by the growing concerns on sustainability and product life cycle of polymer composites, natural fibre with its relatively high specific strength and stiffness, lightweight, and renewable, biodegradable, and low cost manufacture has received significant attention to be developed as an alternative reinforcement in polymer composites [5, 6]. Oil palm is the highest yielding edible oil crop in the world and Malaysia is one of the major oil palm cultivating countries. Palm based lignocellulosic fibres can be extracted from trunk, frond, fruit mesocarp, and empty fruit bunch (EFB). Among the various palm fibre sources, EFB has potential to yield up to 73% fibres and the palm oil industry has to dispose about 1.1 ton of

FIGURE 1: Varying length of the EFB fibres.

EFB per every ton of oil produced [1]. The EFB fibre that is abundantly available as residual produce of the palm oil extraction process was selected in this research study due to its low cost, ease of recyclability, low density, low energy cost, and positive contribution to the global carbon footprint.

Owing to the inherently much weaker mechanical performance of EFB fibre [7, 8], numerous works have been carried out to exploit the potential of integrating more than one type of reinforcement into natural fibres filled composite [9].

Rozman et al. revealed that increasing the volume fraction, V_f , of EFB fibre while reducing the proportion of glass (GLS) fibre leads to decreases in tensile and flexural strengths of polypropylene based hybrid composite [10]. In the work of Karina et al. [1], the authors found that high loadings of EFB fibre in the range of 40–70% V_f into a polyester resin through wet lay-up process significantly improved the flexural strength of the resin by 350%. EFB loadings at 40% V_f displayed similar flexural strength as polyester/GLS fibre composites with much lower densities. While the mechanical properties of hybrid natural fibre composites were well studied, the effect of synthetic fibre length in oil palm EFB hybrid composites remains unexplored. In recent years there has been strong growth in the use of long glass-fibre thermoplastic composite systems in semistructural and engineering applications. The enhanced mechanical performance results from the ability of long fibre to transfer stresses across the fibre-matrix interface as well as a combination of the fibre and matrix properties [11]. Varying length of the fibres affects the capability of interfacial stress transfers and the effects of glass fibre length in PLA-EFB-GLS hybrid fibres composite were investigated in this research work. Glass fibre was chosen as it has been widely used as reinforcement for both thermoplastic and thermosets due to its low cost, reasonable tensile strength and chemical resistance, high dimensional stability, and excellent insulation properties. It is worth mentioning that inclusion of GLS fibre may undermine the environmental impact of the development of natural fibre composites since synthetic fibres generally do not degrade. However, the hybridization of natural fibre with low loading of stronger synthetic fibre can provide avenue to improve the mechanical properties and moisture resistant behaviour of the composites and hence a balance between environmental impact and performance could be achieved.

To yield a more sustainable composite with reasonable mechanical properties and to reduce environmental impact, PLA hybrid composite reinforced with randomly oriented

FIGURE 2: SEM micrograph showing the various diameters of EFB fibres.

oil palm EFB and chopped strand GLS fibres was studied, whereby a large proportion of the ingredients (i.e., PLA, EFB) are harnessed from renewable sources. Homogeneous mixture of the PLA resin and EFB fibres was first prepared using solvent dissolution method and the solidified compound was shredded into pellets. Measured quantity of GLS fibre with predefined length was evenly distributed amongst the PLA-EFB pellets prior to hot compression press into specimen of 1 mm in thickness. The effects of synthetic fibre length, solvent type, and fibre volume fraction on the mechanical properties of the hybrid fibres PLA composite were reported.

2. Experimental

2.1. Materials. The EFB fibres used in this research were supplied by Malaysian Palm Oil Board (MPOB). The fibres have an average length of 4 to 13 mm (Figure 1) and an average diameter of 13.3 to 42.7 μm (Figure 2), giving an aspect ratio in the range of 100 to 10000. The injection grade PLA resin Ingeo biopolymer 3052D was purchased from NatureWorks LLC. The resin was in crystalline form, having specific gravity of 1.24 g/cm³, crystalline melt temperature 145–160°C, glass transition temperature 55–60°C, MFR of 14 g/10 min (210°C, 2.16 kg), and a relative viscosity of 3.3 Pa·s [12]. Chopped E-glass strands of 3 mm and 6 mm length were purchased from JiaXing Sure Composite Co. Chloroform (stabilized with 1% ethanol) and dichloromethane were supplied by EOS Scientific Ltd. under the trademark of RCI Labscan. The solvents used met analytical reagent (AR) grade specifications.

2.2. Material Preparation. The EFB fibres were first washed and sieved under running water to remove sand, mud, and residue from pulverized fibre. The cleaned fibres were subsequently dried at 70°C for a minimum of 24 hours. On the other hand, the PLA pellets were dried in an oven at 50°C for a period of 24 hours to remove moisture content. The conditioned PLA pellets were then weighed into the required masses and sealed by batch into plastic bags containing silica gel to prevent moisture reabsorption prior to solvent dissolution.

TABLE 1: Experimental parameters investigated in this work.

Sample designation*	Volume fraction, V_f (%)		Solvent type	Length of E-glass (GLS) fibre (mm)
	E-glass (GLS) fibre	Empty fruit bunch (EFB) fibre		
CH003	0	20	Chloroform	3
CH053	5	15	Chloroform	3
CH103	10	10	Chloroform	3
CH153	15	5	Chloroform	3
CH203	20	0	Chloroform	3
DC003	0	20	Dichloromethane	3
DC053	5	15	Dichloromethane	3
DC103	10	10	Dichloromethane	3
DC153	15	5	Dichloromethane	3
DC203	20	0	Dichloromethane	3
DC056	5	15	Dichloromethane	6
DC106	10	10	Dichloromethane	6
DC156	15	5	Dichloromethane	6
DC206	20	0	Dichloromethane	6

*Nomenclature for CHXXY and DCXXY:

“CH” and “DC” refer to the solvent type used for the processing (CH = chloroform; DC = dichloromethane); “XX” corresponds to the volume fraction of GLS fibre (ranging from 0 to 20% V_f) while “Y” corresponds to the length of GLS fibre (3 or 6 mm).

2.3. Specimen Fabrication. The dried PLA pellets were dissolved in solvent at a ratio of 60 g resin to 200 mL solvent within a 500 mL Erlenmeyer. The PLA pellets were continuously stirred with a magnetic stirrer for a minimum of 6 hours to ensure complete dissolution. Dried EFB fibres were subsequently introduced into the Erlenmeyer and mixing continues until a homogenous fibre distribution was achieved. The resulting PLA-EFB mixture was then spread out in metal trays precoated with silicone mould release agent and left to dry within a fume hood for a period of 24 hours. The solidified mixture was obtained and shredded into pellets form before being placed in an oven maintained at 60°C for further drying over a period of 24 hours. Figure 3 shows the chopped pellets of the PLA-EFB compound prepared using chloroform as the solvent. E-glass fibres with length of 3 mm and 6 mm were separately sieved to remove foreign particle and to break up clumped fibre. The appropriate masses of GLS fibre were weighed separately by length and sealed into plastic bags.

A stainless steel mould (cavity 200 × 150 × 3 mm) with a 2 mm steel plate insert incorporated was used to produce 1 mm thick specimen sheets. The PLA-EFB pellets and GLS fibres of specific length were mixed and evenly spread into the cavity of the mould. The material was then pressed at 165°C, that is, slightly above the crystalline melt temperature of PLA using a hydraulic hot press. Two different pressure settings, 4.4 MPa and 8 MPa, were employed. The material was then cooled under controlled pressure within the mould prior to demoulding of the solidified specimen plate.

2.4. Experimental Parameters. The effects of fibre loading (ranging from 0 to 20% V_f), the solvent types (chloroform and dichloromethane), and the GLS fibre lengths (3 and 6 mm) on the mechanical properties of the hybrid fibres

FIGURE 3: The chopped solidified PLA-EFB pellets.

filled PLA composite were investigated. Table 1 depicts the experimental parameters used in this work while Figure 4 shows a comparison of the resulting specimen for the EFB: GLS fibre loading of 20:0, 10:10, and 0:20% V_f using chloroform as the solvent type.

2.5. Testing Procedures

2.5.1. Tensile Test. The tensile specimens were milled to ASTM D638-02a Type 1 standard. The specimens were kept in sealed plastic bags with silica gel for a minimum of 24 hours to remove traces of surface moisture prior to testing. All specimens were tested to failure under constant crosshead speed of 5 mm/min using Lloyd Instruments LR50k tensile testing machine. At least three specimens were tested for each formulation.

2.5.2. Flexural Test. The flexural specimens of size 75 × 12.7 mm were prepared with the aid of a vertical bandsaw.

FIGURE 4: Specimens with EFB: GLS fibre loading of 20 : 0, 10 : 10, and 0 : 20% V_f (left to right).

FIGURE 5: Tensile strength of the pure EFB, hybrid EFB-GLS, and pure GLS fibre PLA composite at various % V_f and GLS fibre length produced using chloroform and dichloromethane solvent.

The deflection point was set in accordance with ASTM D790-03 standards and crosshead speed of 1 mm/s was employed in the test. The specimens were sealed into plastic bags with silica gel to remove traces of surface moisture prior to testing. Flexural modulus and flexural strength were calculated based on constitutive equations.

3. Results

3.1. Tensile Properties. Figure 5 depicts the measured tensile properties of the hybrid EFB-GLS fibres filled PLA composite. The hybrid fibres composites showed higher tensile strength compared to composite filled by either EFB or GLS fibres alone, regardless of the solvent type and GLS fibre length. Tensile strength of the hybrid composite was found to increase with GLS fibre loading, that is, from 5 to 15%. Further increase of GLS fibre loading to 20% (0% EFB) resulted in

FIGURE 6: Elongation at break (%) against various % V_f and length of GLS fibre EFB hybrid PLA composite produced using chloroform and dichloromethane solvent.

much lower tensile strength compared to the hybrid EFB-GLS fibres composite for both the 3 mm and 6 mm GLS fibre length. On the other hand, 20% V_f of pure GLS fibre filled composite appeared much stronger in tension than pure EFB fibre filled composite. This can be attributed to the superior mechanical properties of the GLS fibre compared to EFB.

The specimens which were produced using chloroform as the solvent performed significantly better (between 50 and 150%) than the specimens which were produced using dichloromethane. For the specimens with dichloromethane, the 6 mm GLS specimens generally exhibit lower tensile strength compared to the 3 mm GLS specimens (except for specimens with 10% V_f of 6 mm GLS fibre).

The specimens' percentage elongation at break is shown in Figure 6. Slight increase of the elongation at break was observed on specimens with 3 mm GLS fibre up to 15% loading (5% EFB) for both the chloroform and dichloromethane specimens. The elongation at break for the 3 mm GLS fibre then dropped at 20% GLS fibre loading. For the 6 mm GLS fibre specimens with dichloromethane, the elongation at break continues to drop with increasing % V_f of the GLS fibre.

3.2. Flexural Properties. The flexural strength and modulus of the composite subjected to 3-point bending are shown in Figures 7 and 8, respectively. The greatest flexural strength was recorded at moderate loading (15%) of 3 mm GLS fibre with chloroform as the solvent. Both chloroform and dichloromethane specimens at lower loading of 3 mm GLS fibre induced much milder effects on flexural strength. A decrease in flexural strength can be observed for the specimens with 20% 3 mm GLS fibre loading (0% EFB). Composite with 6 mm GLS fibre specimens showed a drop

FIGURE 7: Flexural strength against various V_f and length of GLS fibre EFB hybrid PLA composite produced using chloroform and dichloromethane solvent.

FIGURE 8: Flexural modulus against various V_f and length of GLS fibre EFB hybrid PLA composite produced using chloroform and dichloromethane solvent.

in flexural strength compared to those filled with EFB fibre alone. Varying V_f of the 6 mm GLS fibre from 5% to 20% does not reveal any significant variation in the measured flexural strength.

In terms of the flexural modulus, all specimens showed a drop in flexural modulus from 15% to 20% V_f of GLS fibre

FIGURE 9: The tensile fracture surface of a specimen (15% 3 mm GLS fibre, 5% EFB).

FIGURE 10: The tensile fracture surface of a specimen (5% 3 mm GLS fibre, 15% EFB).

loading. An increasing trend can clearly be seen for hybrid composite with chloroform 3 mm and dichloromethane 6 mm GLS fibre loading up to the 15% V_f . A significant variation in the results of the 3 mm GLS fibre with dichloromethane was observed.

4. Discussion

In general the tensile and flexural tests showed that mechanical properties of the hybrid composite increase with increasing GLS fibre loading, that is, with decreasing concentration of EFB fibres. The result is consistent with existing literature on the performance of oil palm EFB lignocellulosic/thermoplastic composites [13].

A major contributing factor to the strength of fibre reinforced composite could be attributed to the interfacial adhesion between the fibres and the resin matrix [14]. Good interfacial bonds facilitate better stress transfer from the matrix to the fibre [15]. A preliminary examination of the fracture surfaces revealed that significant GLS fibre pull-out occurred, as shown in Figures 9 and 10. The phenomenon implies that GLS fibres and PLA resin failed to establish strong bonding within the specimens, leading to poor load transfer from the resin to the GLS fibres. It is worth mentioning that the current method of observation on fracture surface only provides a fundamental level of understanding. More precise microscopy techniques such as SEM and TEM shall be used to provide more accurate and conclusive remarks.

There are two major factors which could contribute to weak interfacial bonding between the fibre-matrix interfaces: the wetting of the fibres and the fibre distribution within the composite [16]. The EFB fibres were solvent compounded with the resin before being dried and shredded into pellets form. On the other hand, the GLS fibres were introduced and mixed with the shredded EFB-PLA pellets prior to hot compression moulding in order to preserve its length, that

is, 3 mm and 6 mm, respectively. Because the dissolved resin was substantially less viscous than the molten resin, the EFB fibres received far better wetting compared to the GLS fibres. Insufficient GLS fibre wetting would have contributed to the prevalence of GLS fibre pull-out as experienced by the specimens with 20% GLS fibre filled composite (0% EFB). In addition, the GLS fibres tend to agglomerate within the composite specimens, arising due to the preparation procedure where inclusion of GLS fibres took place at the last step of the specimen preparation and it is extremely challenging to achieve a homogenous fibre distribution within the highly viscous molten resin during the hot compression moulding. In particular, longer GLS fibres, that is, those of 6 mm length, experienced greater limitation in achieving uniform mixing and have a higher tendency to agglomerate, which lead to lower efficiency in fibre wetting, hence the lower interfacial bonding and the drop in mechanical properties.

Comparing to the hybrid fibres composite at 15% GLS fibre loading, the recorded lower tensile strength of the homo-GLS fibres PLA composite (i.e., 20% GLS, 0% EFB) may be attributed to the specimen preparation procedure needed to preserve the GLS fibre length, which causes reduced fibres wetting, fibre agglomeration, and poor interfacial adhesion between GLS fibre and PLA matrix interface.

The composite produced with chloroform as the solvent displayed a significantly higher tensile and flexural strength than specimen prepared through dichloromethane. The viscosity of the resin solution with different solvents could have affected the degree of fibre wetting; that is, lower viscosity is more effective in fibres wetting, thereby improving the interfacial bond between the fibres and the matrix. In accordance with the Hansen Solubility Index [17, 18], effective dissolution could be achieved when the solubility parameter of the polymer and solvents is less than 2.5. The solubility parameter of both chloroform and dichloromethane is within this range with that of the PLA. The greater PLA dissolution capability of chloroform could be attributed to its lower surface tension [19] which promotes more effective interchain migration of solvent molecules to induce swelling and dissolution of the PLA resin to yield much lower viscosity. Work done by Byun et al. [20] suggested that the solvent choice would have affected the crystallinity of the PLA although it is unclear whether the same crystallinity is preserved through the compression moulding process.

5. Conclusions

The use of EFB and GLS fibres as reinforcement in a hybrid PLA composite produced results that are outlined as follows.

- (a) The tensile and flexural strength of the hybrid composite generally improved with increased GLS fibre loading, up to 15% V_f , regardless of the solvent type.
- (b) PLA specimens filled with GLS fibres alone (20% GLS, 0% EFB) depict poorer mechanical properties compared to EFB-GLS fibres hybrid PLA composites.
- (c) The use of chloroform to compound EFB fibres in PLA resin improved the tensile and flexural

properties of the specimens by 50 to 150% compared to dichloromethane.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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