

# ECO-BIOCOMPOSITE MATERIALS FOR SHOCK CUSHIONING APPLICATION: AN OUTLOOK OF THE POTENTIALS AND CHALLENGES

David, N. V.\*, Siti Syahidah M. Rum

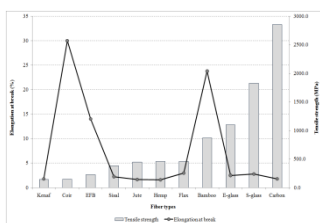
Faculty of Mechanical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

\*Corresponding author  
davidfkm@salam.uitm.edu.my

## Article history

Received  
5 December 2014  
Received in revised form  
30 April 2015  
Accepted  
9 June 2015

## Graphical abstract



## Abstract

Recently there is an increased concern for the development of not only well-designed but also sustainable materials. Material sustainability is associated with the extraction of renewable resources and disposal procedures that would not injure our ecosystem. In this respect, indigenous agricultural wastes or by-products are suitable alternatives to lightweight and disposable green-materials at low cost. Agro-waste and biomass materials are plentiful in many parts of the world including Malaysia. Fibers obtained from agricultural by-products are often used as fillers or reinforcement in non-biodegradable polymer matrix. Polylactic acid (PLA), which is a compostable and biodegradable thermoplastic, is derived from renewable agro-sources such as potato, corn, or sugarcane. The mechanical and thermal properties of select biofibers-filled PLA composites are comparable to that of the composites made from conventional fibers. Research findings imply the feasibility of processing PLA with natural fibers such as kenaf using existing manufacturing technologies. Natural fiber filled biodegradable polymer composite materials have the advantage of simple and safe disposal over petroleum-based polymers besides generating new low-carbon economy for the plantation sector. However, research outcomes show that the fiber/matrix interface of PLA and natural fiber is weak due to incompatible surface properties of the two material types. In this article, issues pertaining to fiber/matrix interfacial adhesion, potential renewable sources of polymers and processing technologies of natural fiber (or -eco)-biocomposite materials are reviewed. The prospect of replacing traditional polymers obtained from non-renewable fossil resources with biopolymers to develop sustainable eco-biocomposite materials for shock cushioning application such as for packing and packaging materials is discussed in particular.

Keywords: Biopolymer; natural fiber; environment; shock cushioning; packaging materials

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## 1.0 INTRODUCTION

Advancement in materials science and engineering during the past 20 years were driven mainly by the need to produce lightweight materials that are comparable in performance to their contemporaries. There is recently an increased concern for the development of not only well-designed but also sustainable materials. Material sustainability is associated with the extraction of renewable resources and disposal procedures that would not injure our ecosystem. In this respect, indigenous agricultural wastes or by-products are

suitable alternatives to lightweight and disposable green-materials at low cost.

Natural resources and thereby agro-waste and biomass materials are plentiful in many parts of the world including Malaysia. For example, a recent report estimated an overall oil palm empty fruit bunch production of 7.29 million tonnes (by dry weight) from the Malaysian oil palm industry in 2009 [1]. It is to be noted that no chemicals are usually involved in the production of oil palm fibers. Other sources of natural fibers include inedible crop wastes such as coir, sunflower stalk, rice husk and kenaf, which is regarded as an industrial crop in

Malaysia, are lower in density, safer to handle and cheaper than inorganic fibers, and can be processed using existing polymer manufacturing technologies. Agro-based fibers are often used as fillers or reinforcement in polymer matrices that are derived from non-renewable petroleum resource.

Poly(lactic acid) (PLA), which is a compostable and biodegradable thermoplastic, is derived from renewable agro-sources such as potato, corn, or sugarcane. PLA exhibits mechanical behavior that is comparable to other thermoplastics including polyethylene and polystyrene [2]. Films made from PLA are fast becoming a material of choice in food packaging in the United States of America and Canada [3].

Research findings indicate the feasibility of processing PLA with natural fibers such as kenaf using existing manufacturing technologies [4]. Natural fiber filled biodegradable polymer composite materials have the advantage of simple and safe disposal over petroleum-based polymers [5] besides generating new low-carbon economy for the plantation sector. The mechanical and thermal properties of select biofibers-filled PLA composites are comparable to that of the composites made from conventional fibers [6-9]. However, research outcomes show that the fiber/matrix interface of PLA and natural fiber is weak due to incompatible surface properties of the two material types [7,8,10]. This warrants further research to optimize the interface compatibility of the fiber and the biopolymer, e.g., PLA.

This article is intended to communicate recent information and issues pertaining to potential renewable sources of synthetic polymers, processing technologies of natural fiber (or -eco)-biocomposite materials and fiber/matrix interfacial adhesion. The prospect of replacing traditional polymers obtained from non-renewable fossil resources with biopolymers to develop sustainable eco-biocomposite materials for shock cushioning application such as for packing and packaging materials is discussed in particular.

## 2.0 BIODEGRADABLE POLYMERS

### 2.1 Types and Availability

Major classes of biodegradable synthetic polymers (hereon called biodegradable polymers) include polyesters and their copolymers, polyurethanes (PU), polycarbonates, polyesteramides and polymer systems based on polypropylene-fumarates, PU and acrylate/urethane. Interested readers may refer to [11] for a detailed review of these families and other classes of biodegradable polymers.

The review presented here will focus on polylactic acid (PLA), which is a commercially available biodegradable thermoplastic. PLA is usually derived from renewable agro-sources such as potato, corn starch, or sugarcane. The worldwide consumption of biodegradable polymers (also called bio-plastics) like PLA has grown nearly 600% between 2000 and 2008 [12]. The 2010 global production of bio-plastics is about 400,000 tonnes and expected to rise to 2.1 million tonnes in the next three years [12].

### 2.2 Processing and Properties of Biodegradable Polymers

The structural, thermal, crystallization, and rheological properties of PLA are reviewed in relation to its conversion to molded parts, films, foams, and fibers in [4]. PLA can be processed on standard converting equipment including

extrusion, injection molding, blown film and foaming. The physical properties of PLA can be tailored in batch foam processing by producing foams with reduced cell size, increased cell density and lowered bulk foam density [13]. These foams would be ideal for shock cushioning applications such as in the packaging and packing industries. However, it has been reported that the range of mechanical properties that can be obtained using different copolymerization methods to address the needs of different applications is still limited [11].

### 2.3 Current Applications and Market Potential

Biodegradable polyesters such as polyglycolides and PLA are used in the production of disposable clinical products such as surgical sutures and implants [11,14]. Injectable polymers based on urethane and urethane/acrylate have been utilized to develop delivery systems for tissue engineered products and therapies [11].

Biodegradable polymers are also being used to produce packaging materials. There are about 45 worldwide competitors in the bio-based packaging sector [15]. Hitherto, there is no protective packaging company in Malaysia that produces bio-based packaging materials [1]. Two commercially available or research-level products that are comparable only in terms of green material concept are listed in Table 1.

**Table 1** Biodegradable materials for packaging application

Features	EcoCradle™ Mushroom® <sup>a</sup>	PSM® (HL-300) <sup>b</sup>
Country of origin	USA	Hong Kong
Product launched in	2010	2005
Market size	Still growing the USA	Covers all major industrial nations
Malaysian agent / reseller	Not available	Adikem Resources Sdn. Bhd.
Base green material	Mushroom (fungal mycelium)	Corn starch
Cost of material	USD\$2.60 per kg	USD\$1.00 per 16-oz box
Selling price	USD\$0.75 – USD\$5.00 per part (comparable to traditional EPS)	Data not available
Processing method	Specialised production 'grow trays' (mould) in which the mushroom grows to the required dimensions and shape	Traditional methods including injection moulding and thermoforming

<sup>a</sup> [http://www.naturalnews.com/029463\\_mushrooms\\_packing\\_material.html](http://www.naturalnews.com/029463_mushrooms_packing_material.html) [Retrieved 10/15/2014].

<sup>b</sup> <http://www.psm-hk.com/Eng/material.htm> [Retrieved 10/15/2014].

In 2005, the total market value of global packaging industry is USD\$425 billion out of which Asia has a 27% share worth USD\$114 billion [6]. The sales of packaging products is experiencing an average annual growth rate of 3.5% since 2005 and are expected to hit USD\$740 billion in 2014 [5]. The global value of bio-packaging materials in 2010 is estimated to be USD\$1.6 billion.

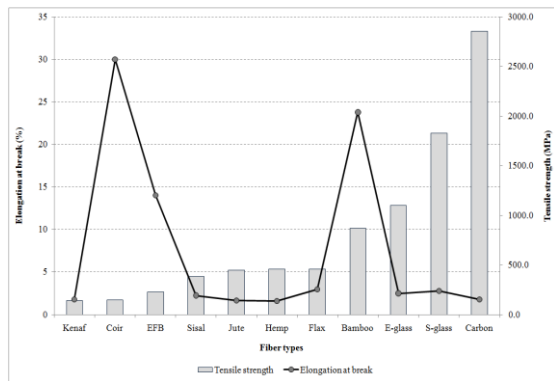
### 3.0 NATURAL FIBERS

#### 3.1 Type, Properties and Availability

Natural fibers (NF) or biofibers such as the flax, hemp, coir, bamboo, kenaf and oil palm empty fruit bunch (EFB) fibers are naturally occurring cellulosic composites made of the crystalline microfibril reinforcement in an amorphous lignin and hemicellulose matrix [16]. The fibril microstructure scales ten to thirty nanometers. The cellulose, hemicellulose and lignin contents of a NF influence its mechanical properties [17]. Amongst the NFs mentioned above, kenaf and EFB will be emphasized in the following discussion.

Cellulose is one of the stiffest and strongest constituents in a NF and thus is the main structural component of the NF. It provides strength and structural stability to the NF. It is reported in [18] that the tensile properties of NFs generally increase with the increase of cellulose content of the fibers. In kenaf, for example, the cellulose content of the core (center pith-to-bast) fiber and the bast (epidermis skin-to-outer core) fiber is about 50.6 % [19] and 60.8% [20], respectively. Interestingly, kenaf comprises around 60 wt% core fibers and 40 wt% bast fibers. A strand of EFB has a total cellulose content of about 70% [21].

NFs generally possess high failure strain despite their relatively poor strength and excellent specific properties due to their lower densities. Figure 1 shows the comparison of mechanical properties of select types of natural and conventional synthetic fibers.



**Figure 1** Comparison of mechanical properties of select types of natural and conventional synthetic fibers

As shown in Figure 1, biofibers inherently have a higher degree of variability compared to synthetic fibers, i.e., low homogenization and non-uniformity of properties although of comparable structural and biological make-up. Flax fibers possess high specific properties while coir fiber has the lowest density and highest elongation to failure among a range of NFs. Besides its relatively better mechanical properties, the hollow microstructure of bamboo fibers permits excellent heat dissipation and ventilation [22].

Malaysia is endowed with abundant natural resources of biomass and agro-waste materials. Among others, oil palm empty fruit bunch (EFB), which is a by-product from the oil palm industry, is one of the largely available agro-waste resources in Malaysia. A recent report estimated an overall oil palm empty fruit bunch production of 7.29 million tonnes (by dry weight) from the Malaysian oil palm industry [1]. The palm oil industry generates

approximately 94% of the biomass feedstock in Malaysia [1]. In proportion to this, the Malaysian Economic Transformation Programme roadmap estimates a growth of about 247% of the palm oil industry in terms of Gross National Income contribution by the year 2020. Table 2 lists the types of NFs and their availability.

#### 3.2 Current Applications and Market Potential

The advantages of NFs include availability, low cost, environmental friendly, low specific density, deformability, less abrasive to processing equipment, manufacturability, carbon-dioxide neutral and energy efficient production. These benefits are central to the possible utilization of NFs either on its own or as reinforcement or filling agent in composites for a variety of applications.

**Table 2** Fiber types, production and exporting countries [23]

Fiber type	Annual global production (10 <sup>3</sup> tonnes)	Some of the main exporters
Coir	100	India, Philippines, Malaysia
Hemp	215	Yugoslavia, China
Kenaf	770	Iraq, Tanzania, South Africa
Flax	810	Borneo
Bamboo	10000	China
EFB	10000 <sup>a</sup>	Malaysia, Indonesia

<sup>a</sup><http://www.probos.net/biomassa-upstream/pdf/reportBUSA36.pdf> [Retrieved August 5, 2011].

The utilization of NF in a woven architecture for body armor application is studied in [24]. It was found there that flax composites exhibited better energy absorption than hemp and jute composites. Fabrics made from 100% bamboo fibers are good at sweat absorbing (due to its hollow microstructure as mentioned above), possess natural anti-UV properties and high failure strain.

A number of scientific and developmental studies have been carried out on the utilization of EFB, such as in particle boards (e.g., [25]), medium density fiberboard (e.g., [26]), pulp (e.g., [27]), and sound absorption panel (e.g., [28]). The utilization of EFB as renewable fuel after its conversion to biodiesel is noteworthy here [1]. An advantage of EFB over palm oil or other biomaterials such as corn starch, for the production of biodiesel is that the fuel is made of essentially inedible raw material and therefore does not tap into the human food reservoirs.

Besides the applications stated above, EFB has been also used as reinforcement (in form of short or chopped fibers) or filler (in particulate form) in conventional polymer composites for various other applications (e.g., [29-32]). The classification of composites into hybrid biocomposites, textile biocomposites and green composites is discussed in [33].

Among the challenges of using NFs including EFB as a means of reinforcing polymer-based composites are their high moisture content (due to hydrophilic surface nature), material heterogeneity and non-uniformity of mechanical and physical properties. Some of the issues pertaining to these challenges are discussed next.

#### 3.3 Processing Issues Related to Natural Fiber/Conventional Polymer Composites

The surface of NFs contains water-attracting (hydrophilic) hydroxyl (OH<sup>-</sup>) group. Thermoplastic polymers such as

polyolefin or polyurethane normally used as the matrix in filled or reinforced polymer composites are water-resisting (hydrophobic). This poses the dual problem of composite fabrication, i.e., blending the two constituents with the opposing hydro-features stated above, and the resulting mechanical properties that are influenced profoundly by the fiber/matrix surface compatibility.

As a measure to overcome this challenge, the surface of the NFs are generally treated or modified to attune their surface characteristics favorably to that of the polymer matrix. Chemical surface modifiers or coupling agents such as maleated polypropylene (MAPP) and polyethylene glycol [34] can be employed for this purpose. For example, it was reported in [35] that the flexural and impact properties of EFB filled polypropylene (PP) composites are enhanced by adding the MAPP to the EFB-PP blend. The anhydride group in MAPP couples the fiber to the matrix and improves the transfer of applied stress from the matrix to the fiber.

In another study, two types of coupling agents, namely, Epolene (E-43-amaleic anhydride-modified PP), which was also used in [36], and 3-(trimethoxysilyl)-propylmethacrylate (TPM) were shown to improve the tensile strength of a hybrid composite consisting of EFB and glass fiber in PP matrix [37]. The properties of polyurethane (PU) composites reinforced by fibres obtained from oil palm resources are reported in [37-38]. Packaging material made from bio-based PU has shown tremendous improvement in terms of the mechanical properties of the composites.

The incorporation of different EFB fibre sizes at different levels of concentration namely, 5.5 wt% of 45 – 56  $\mu\text{m}$ , 4.5 wt% of 100 – 160  $\mu\text{m}$  and 2.5 wt% of 200 – 315  $\mu\text{m}$  in the PU foam produced were conducted in the study. The results indicated that the compressive strength of the PU/EFB foam composite increased by 11 to 30%. Furthermore, other properties of the PU/EFB foam composite such as the tack free time, density, percentage of open cell, relative energy absorption, compressive stress, and tear strength showed that oil palm-based foams are suitable for packaging and shock-absorption applications.

#### 4.0 PROPERTIES AND PERFORMANCE OF ECO-BIOCOMPOSITES

The mechanical, impact and thermal properties of NF-filled or reinforced biodegradable polymer composite (or eco-biocomposite) materials can be tailored (e.g., [39-40]) using suitable NF type alongside proper surface modifiers or coupling agents in tandem with optimal fiber content.

For instance, the impact and tensile properties Cordenka rayon fibers and flax fibers reinforced PLA composites determined in [41] indicate that the Cordenka fibers provided the highest impact strength and tensile strength at a loading of 30 wt%. On the other hand, the greatest tensile modulus was obtained with the flax fibers.

Natural flax fibers as the reinforcement in PLA matrix was investigated in [6]. It was found there that addition of 30 wt% of flax improved the tensile strength of the PLA/flax composite by 50% compared to PP/flax composites with similar fiber loading. It has to be noted that the PLA/flax composites were prepared using conventional twin-screw extrusion and compression molding processes. No degradation of PLA was observed

due to the compounding process or inclusion of the flax fibers. The thermal properties of plain PLA were also improved with the addition of the flax fibers.

#### 4.1 Fiber/Matrix Interfacial Adhesion

Eco-biocomposites comprised of kenaf fiber reinforced PLA are fabricated by carding followed by treatment with a 3-glycidoxypropyl trimethoxy silane and hot-pressing in [42]. The effects of the silane coupling agent on composite properties was increased tensile modulus and heat deflection temperatures as well as reduced swelling in water. Mechanical properties of the eco-biocomposites at temperatures past the glass transition are improved in comparison to the 100% PLA polymer. Thermal properties such as the glass transition, melting temperature, and crystallinity of the PLA matrix are unaffected by incorporation of kenaf into the composites. Interfacial adhesion between NF and PLA can be characterized at the micro-scale using microbond test [e.g., 43].

#### 4.2 Biodegradability of Eco-Biocomposites

Sustainable eco-biocomposites are expected to satisfy a few requirements including renewable and/or recycled resources should be utilized for their manufacture, the processing operations should be benign and energy effective, no hazardous environmental effects should arise during any stage of their life cycle, and waste management options should be implemented [44].

For example, the biodegradability of kenaf/PLA composites was examined for four weeks using a garbage-processing machine in [45]. It was found that the weight of composites decreased 38% after four weeks of composting. Eco-biocomposite materials thus have the advantage of simple and safe disposal over petroleum-based polymers (e.g., [5], [46]) besides generating new low-carbon economy for the plantation sector.

#### 5.0 ECO-BIOCOMPOSITES FOR PACKAGING APPLICATIONS

Packaging materials (including packing materials) for logistical purposes are designed to protect electrical and electronic products and other fragile consumer goods from damages due to shock and/or excessive vibrations during handling and transportation. Some packing materials are employed just as space-filler to avoid content mobility without useful protection against shock and vibrations. At present, the predominant packaging materials in the market are made from expanded polystyrene (EPS) and polyethylene. This fully polymeric material is not expensive when produced in mass quantity but the disposal of the non-biodegradable and photolysis-resistant EPS often causes environmental and health concerns. It has been reported in the literature that the production of petroleum-based packing foams like EPS releases ten times the volume of carbon dioxide and consumes up to eight times the energy than that needed to produce a sample of mushroom-based packing material [12].

As mentioned in Section III-B above, agro-waste materials have been studied and commercially produced in forms of particle boards, medium density fiberboards, sound absorption panels and structural members for a variety of relevant applications. The authors are currently



pursuing the idea of producing packed agro-fibers (including coir and kenaf) that are cemented in a biodegradable PLA polymer matrix for shock cushioning and vibration protection applications.

The utilization of oil palm biomass and coir (agricultural bio-waste) and kenaf (trade crop) for applications such as shock and vibration protection adds commercial value to the existing worth of these new commodities as the demand volume for such bio-packaging increases in future. The successful commercialization of the prototype would create a new local green protective packaging industry that shares the fortune with the plantation workforce. A less energy intensive low-carbon economy of downstream plantation activities is expected to balance the steady but sure depletion of nonrenewable resources now being exploited for many including packaging purposes.

## 6.0 CONCLUSION

A general review of critical information and issues pertaining to potential renewable sources of synthetic polymers, processing technologies of eco-biocomposite materials, fiber/matrix interfacial adhesion is presented. The mechanical, impact and thermal properties of NF-filled or NF-reinforced biodegradable polymer composite (or eco-biocomposite) materials can be tailored using suitable NF type alongside proper surface modifiers or coupling agents in tandem with optimal fiber content. The prospect of replacing traditional polymers obtained from non-renewable fossil resources with biopolymers such as PLA to develop sustainable eco-biocomposite materials for shock cushioning application such as for packing and packaging materials is discussed in particular. The utilization of agricultural bio-waste for applications such as shock and vibration protection adds commercial value to the existing worth of these commodities as the demand volume for such bio-packaging increases in future.

## Acknowledgment

The work reported here is jointly funded by MOSTI Malaysia and the Ministry of Education Malaysia under the auspices of the ScienceFund and Research Acculturation Collaborative Effort (RACE) grants, respectively. This support is gratefully acknowledged. The Research Management Center of Universiti Teknologi MARA (UiTM) Malaysia is thanked for its administration and management services of the grants.

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