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Md Rezaur Rahman *Editor*

Acacia Wood Bio-composites

Towards Bio-Sustainability of the Environment

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Editor
Md Rezaur Rahman
Faculty of Engineering
Universiti Malaysia Sarawak
Kota Samarahan, Sarawak, Malaysia

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Contents

Introduction of Various Types of Acacia Wood

Md Rezaur Rahman, A. Kakar, S. Hamdan, M. K. B. Bakri, N. Julai and P. L. Nyuk Khui

Abstract Genus Acacia has a complex history in globally, there are categorize them as plants, shrubs, and large trees. This chapter covers various types of acacia species, and plantation of Acacia trees in regions such as Malaysia and Portugal. Some Acacia species are valued while others are considered as an invasive species towards the surrounding region of flora. Numerous methods were applied to control such invasive species, one of the alternative method of control is the application of acacia wood as timber product for wood industries. Heartwood and sapwood content are one of the important features in timber, as the content affects the quality, durability and susceptibility of the product which is attack by fungi. Some studies reported a relatively high heartwood content in acacia woods in acacia tree plantations, hence acacia timber has potential as a desirable wood working material and other commercial or industrial applications.

Keywords Types · Acacia · Wood · Properties

1 Introduction

The genus, Acacia, includes a group of plants. The categorization of Acacia has a very complex history. Acacia was first termed by Philip Miller, in the year 1754. However, the definition of this genus was changed less than a century later, in 1842, by George Bentham. George Bentham restricted this group's recognition to mimosoid plants, which characteristically have numerous free stamens (the male fertilizing organ of a flower). As of Maslin et al. (2003) recognized that there were more than 1350 species of Acacia. The classification and recognition of the types of Acacia have been so controversial through history, that it was explicitly mentioned in a journal Retypification of Acacia, in 2011 (Leary et al. 2006). Thiele et al. (2011) clearly discussed

Md R. Rahman (✉) · S. Hamdan · M. K. B. Bakri · N. Julai · P. L. Nyuk Khui
Faculty of Engineering, Universiti Malaysia Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

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Table 1 Continental distribution of each Acacia genera (Thiele et al. 2011)

Post-Vienna names	Asia	Australia	America	Africa	Total
Acacia	32	9	52	83	176
Senegalia	48	2	79	74	203
Acaciella	0	0	15	0	15
Mariosousa	0	0	13	0	13
Racosperma	12	1017	0	1	1030
Total	92	1028	159	158	1437

the controversial issues and recognized a total of 1437 acacia species. However, the number mentioned by Maslin et al. (2003) was iterated by the Australian government, on their Australian National Botanical Garden website (in collaboration with Centre for Australian National Biodiversity Research), which was updated in 2015 (Maslin et al. 2003). Although the discrepancies are much smaller now, there still exists some debate over the category of Acacia. To keep the data as factually correct as possible, this chapter will focus on the following major types of Acacia that have the latest recognition by the scientific community at large. These five genera belong to the Post-Vienna controversy, namely: Acacia, Senegalia, Acaciella, Mariosousa and Racosperma. Table 1 shows the distribution of these species throughout the world.

2 Genus Acacia

According to some studies the genus Acacia is the 2nd largest genus within the Leguminosae, where 1200–1352 species were identified (Leary et al. 2006; Maslin et al. 2003, 2013). The genus Acacia includes small shrubs, lianas and large trees and it is found in a large range of environments, from dry deserts to mountainous forests. There are totally 176 species under the genus Acacia. Of which, 83 are found in Africa and 64 of them are found in the five eastern African countries: Rwanda, Uganda, Kenya, Burundi and Tanzania. Whereas, in Australia, Acacia is referred to as Wattle. Although in 2010 there were 1020 truly recognized species of genus Acacia, today there are 1028 species (Thiele et al. 2011; González-Orozco et al. 2011). Although none of the species under the genus Acacia have been listed under the globally threatened plant species, some of them are under the Red List process by the IUCN. Most of them are vulnerable due to human activities such as burning, agricultural encroachment, charcoal production and livestock damage. Since they are not protected by law, they were given little importance as far as regional biodiversity assessments were concerned (Marshall et al. 2012) (Figs. 1 and 2).

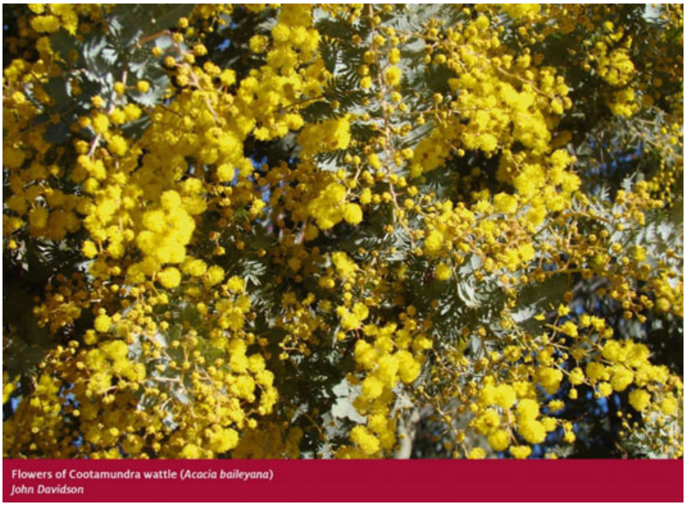

Fig. 1 Genus Acacia (Acacia Forest 2019)

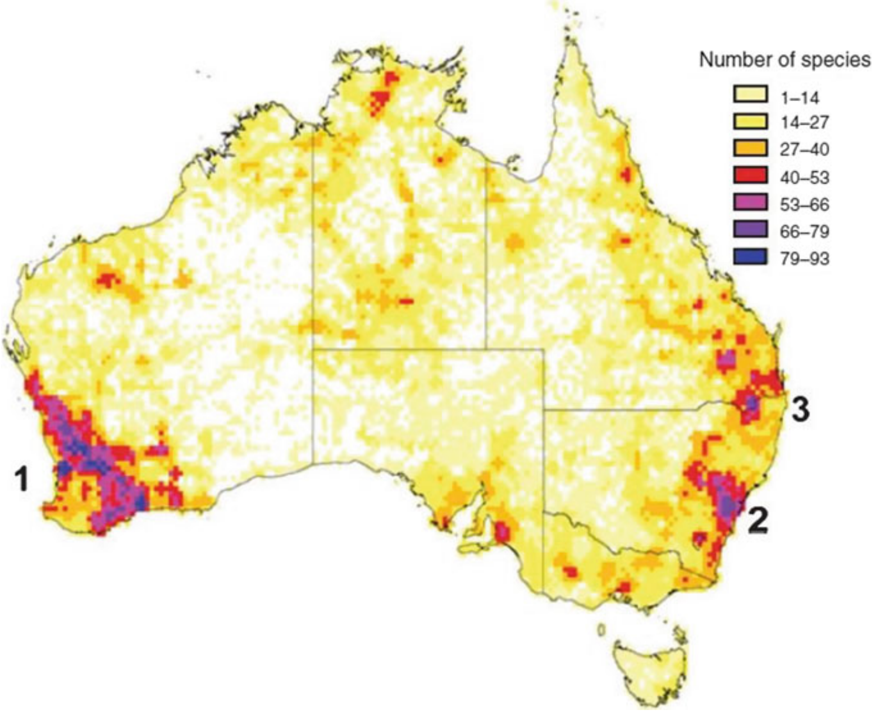

Fig. 2 Distribution of genus Acacia in Australia (González-Orozco et al. 2011)

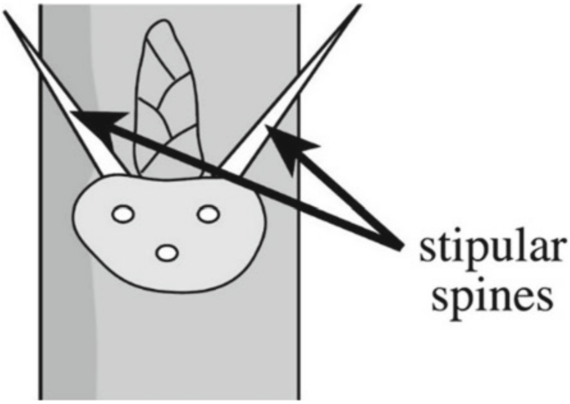

Fig. 3 Stipular spines (Simpson 2010)

3 Genus Senegalia

It is primarily found in Asia, South America and Africa. These shrubs and trees can be found in a broad geographical range, from the Mediterranean to South Africa north. Their growth is restricted to semi-desert scrub and dry savannas habitats (Acacia Forest 2019). It is often confused with Vachellia, however there are few key differences. Senegalia is distinguished by certain macromorphological characteristics such as the type and presence of stipular spines and/or prickles. Senegalia may or may not have prickles, however, they never have stipular spines, unlike Vachellia. Spines are defined as the modified stipules or parts of leaves, for example, extensions of leaf veins. Whereas, prickles are the pointed growth from epidermal layer in the stem and do not emerge from the nodes (Figs. 3, 4 and 5).

When Senegalia has prickles, they are usually dispersed, although on occasions they may be grouped in pairs or threes. Additionally, their pollen is porous and lacks columellae. In contrast, Vachellia have colporate pollen as well as columellae (apertures that include both, rounded pore and groove). In contrast with the Vachellia, Senegalia do not exhibit inflorescence, instead, they spicate or capitate.

Although macro-morphological characteristics are considered important in defining major groups of Acacia, as done by Maslin et al. (2003) and in spite of their major macro- and micro-morphological differences between Senegalia and Vachellia (Table 2), clearly defining the monophyletic lineages within these genera is extremely difficult when based merely on their morphological traits (Miller et al. 2003).

4 Genus Acaciella

Acaciella is typically found in Mexico, but it has also been introduced to India, Brazil, Indonesia, the Dominican Republic, Australia, Papua New Guinea, Thailand, the Philippines, and Pakistan. They do not to possess harmful defense mechanisms

Fig. 4 Acacia prickles

Fig. 5 Acacia prickles

Table 2 Identifying Senegalia

Character	Senegalia	Vachellia
Involucre on peduncle	Absent	Present
Stipular spines	Absent	Present
Prickles	Present	Absent
Pollen collumelae	Absent	Present
Pollen aperture type	Porate	Colporate

unlike the Acacia genus. Among the 15 species, *Acaciella angustissima* is the most studies species of this genus. Initially, in 1768, *Acacia angustissima* was known as *Mimosa angustissima*. However, in 1896, Kuntze transferred it to the Acacia genus. It was therefore known as *Acacia angustissima* until 2006, although Britton and Rose had proposed its transfer to Acaciella in 1928. In 2006, the taxonomic review by Rico Arce and Bachman persuaded the change to Acaciella and therefore it is now known as *Acaciella angustissima* (Csurhes et al. 2016) (Fig. 6).

Acaciella angustissima is infamous for being highly morphologically variable, as it has misled several taxonomists to misidentify species that share similar characteristics. For example, the size of its extended branches may vary from being completely smooth to those with short hair-like structures (Fig. 7). Similarly, its leaves and flowers vary greatly, making it very difficult to identify the species with a reasonable doubt, based on its macro-characteristics (Figs. 8, 9, 10 and 11).

Acaciella angustissima thrives in an infertile, free-draining, acidic soils in its native range (including black and red acid soils). However, it may also be grown on a wide range of soils, including vertisols of slightly alkaline pH. Currently, it is used mainly for grazing cattle (Csurhes et al. 2016). M. de L. Rico Arce and S. Bachman Herbarium (2006) reported that the wood of *Acaciella angustissima* is perceived to be weak because its branches tended to stoop and break even with slight winds (Seigler and Ebinger 2006). However, it lacks experimental testing and information about

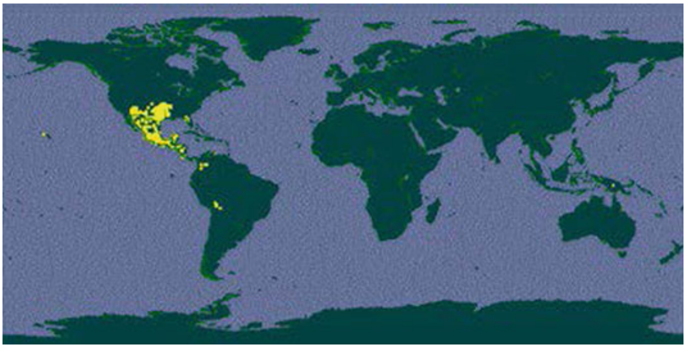

Fig. 6 Worldwide distribution of Aciaciella. Yellow depicts highest density, light green indicates sparse growth and dark green signifies absence of Acaciella

Fig. 7 Stem with fine hair-like structures. *Photo* Sheldon Navie (Csurhes et al. 2016)

Fig. 8 Flower clusters of *Acaciella angustissima*. *Photo* Chris Gardiner (Csurhes et al. 2016)

its physical and mechanical properties post-heat, chemical and surface treatments, which discourages its use as a raw material for manufacturing goods (Table 3).

5 Genus Mariosousa

The species under genus Mariosousa grow only in tropical and subtropical regions of Central America, Mexico and the southwestern United States. Apart from their close geographic locations, their morphological characteristics and molecular data aid in identifying this distinct genus and it is known as the *Acacia coulteri* group. One of their most obvious distinctions include the lack of prickles and lianas (Seigler et al.

Fig. 9 Immature fruit of *Acaciella angustissima*. *Photo* Sheldon Navie (Csurhes et al. 2016)

Fig. 10 Mature fruit of *Acaciella angustissima*. *Photo* Sheldon Navie (Csurhes et al. 2016)

2008). The species belonging to genus Mariosousa are known to be stronger than *Acaciella angustissima*. However, this is known primarily based on observations and physical details recorded by taxonomists rather than laboratory testing. Being a new genus in Acacia, it did not receive much attention from researchers until very recently (Fig. 12; Table 4).

6 Genus Racosperma

The genus Racosperma was listed under Acacia in 1829, after a publication by C. F. P. Martius. By the year 2003, 967 species were defined under the genus Racosperma. Of which, 948 were found in Australia, 10 in Asia (including 7 found in Australia),

Fig. 11 Close-up of the seeds of *Acaciella angustissima*. *Photo* Sheldon Navie (Csurhes et al. 2016)

Table 3 Acaciella species and their conservation categories (Seigler and Ebinger 2006)

Acaciella species	Conservation
A. angustissima (Mill.) Britton & Rose var. *augstissima*	Least concern (LC)
A. angustissima var. *filicioides* (Cav.) L. Rico	Least concern (LC)
A. angustissima var. *texensis* (Nutt. Ex Torrey & A. Gray) L. Rico	Least concern (LC)
A. barranracana (H. Gentry) L. Rico	Data deficient (DD)
A. bicolor Britton & Rose	Vulnerable (VU)
A. chamelensis (L. Rico) L. Rico	Vulnerable (VU)
A. glauca (L.) L. Rico	Least concern (LC)
A. goldmanii Britton & Rose	Least concern (LC)
A. hartwegii (Benth.) Britton & Rose	Near threatened (NT)
A. igualensis Britton & Rose	Least concern (LC)
A. lemmonii (Rose) L. Rose	Near threatened (NT)
A. painteri Britton & Rose var. *painteri*	Near threatened (NT)
A. painteri var. *houghii* (Britton & Rose) L. Rico	Least concern (LC)
A. rosei (Standi.) L. Rico	Least concern (LC)
A. sotoi L. Rico	Critically endangered (CR)
A. sousae (L. Rico) L. Rico	Vulnerable (VU)
A. tequilana (S. Wats.) Britton & Rose var. *tequilana*	Least concern (LC)
A. tequilana var. *crinita* (Rose) L. Rico	Near threatened (NT)
A. tequilana var. *pubifoliolata* L. Rico	Endangered (EN)
A. villosa (Sw.) Britton & Rose	Least concern (LC)

Fig. 12 *Mariosousa acatlensis. Photographer* B. R. Maslin Bark (Seigler and Ebinger 2006)

2 in Madagascar and the Mascarene island and 7 in the Pacific region (Maslin et al. 2003). However, a study in 2011 revealed few changes in the recognized species, as they found 12 in Asia, 1017 in Australia and only 1 in Africa (Thiele et al. 2011). This makes genus Racosperma the most abundant genus (under Acacia) in Australia and the world. However, in spite of this abundance, there is little information available on this genus other than its taxonomy history.

7 Acacia as Invaders and Potential Resource

Many acacia species have been planted throughout the world. Some are valued, while others are viewed as invasive to the ecosystem. Genus Acacia, genus Acaciella and genus Racosperma have all been labeled as invasive by various researchers (Csurhes et al. 2016; Lorenzo et al. 2009; Forrester et al. 2010; Impson and Hoffmann 2019; Yelenik et al. 2004; Richardson and Rejmánek 2011). They are considered to be

Table 4 Genus Mariousa's species and associated taxanomists (Seigler and Ebinger 2006; Seigler et al. 2008)

Species	Taxanomist
Mariosousa acatlensis	(Benth.) Seigler & Ebinger
Mariosousa centralis	(Britton & Rose) Seigler & Ebinger
Mariosousa compacta	(Rose) Seigler & Ebinger
Mariosousa coulteri	(Benth.) Seigler & Ebinger
Mariosousa dolichostachya	(S.F. Blake) Seigler & Ebinger
Mariosousa durangensis	(Britton & Rose) Seigler & Ebinger
Mariosousa mammifera	(Schltdl.) Seigler & Ebinger
Mariosousa millefolia	(S. Watson) Seigler & Ebinger
Mariosousa russelliana	(Britton & Rose) Seigler & Ebinger
Mariosousa salazarii	(Britton & Rose) Seigler & Ebinger
Mariosousa sericea	(Martens & Galeotii) Seigler & Ebinger
Mariosousa usumacintensis	(Lundell) Seigler & Ebinger
Mariosousa willardiana	(Rose) Seigler & Ebinger

invasive because they tend to consume natural resources of their new environment, causing the native habitat to suffer. This consumption may include but may not be limited to water, minerals, nitrogen in the soil, sunlight or even land by spreading like weed. This has created concerns for many countries and ecologists, who are trying to limit the growth and spread of the invasive species that are destroying the lives of the native flora and fauna (Wilson et al. 2011; Richardson and Rejmánek 2011). Many techniques have been tried to get rid of the invaders, such as seed-destruction, however, based on hard data, such attempts have failed (Impson and Hoffmann 2019; Sitzia et al. 2015).

With growing concerns, and failed attempts with expensive destruction methods, researchers have now considered using Acacia as a resource for wood in wood composites for commercial use (Wilson et al. 2011; Rahaman et al. 2012). This would not only make destroying invaders easy, but also provide for cheaper products. However, this does not imply that all the invasive species may be used as a source of raw material. In order to use them most efficiently several factors need to be considered. Wilson et al. (2011) have published a study on risk assessment, eradication and biological control. In their study they consider various factors including invasiveness potential, biological and ecological traits, responsible utilization, public awareness, global and local co-operation, factors that impede eradication, etc. (Wilson et al. 2011).

Considering all such complex factors is important in order to maximize the benefits and minimize any future harm that may be caused by harnessing invaders as resource for raw materials. Through the platform provided in this study, this book aims to cover various treatments, tests and other engineering tools that may help to convert undesirable waste into valuable resource, while keeping in mind the impact it may have on its environment.

8 *Acacia mangium* Willd. Evergreen Tree

There are numerous countries such that has established acacia wood plantations with the purpose of producing acacia wood for numerous applications. An example of the type of fast-growing acacia tree used for these established plantations is the evergreen tree; *Acacia mangium* Willd. The *Acacia mangium* evergreen tree falls under the genus acacia genera, in which is classes in the Phyllodinae subgenus of seven sections comprises of more than 900 species (Hedge et al. 2013). In addition, *Acacia mangium* is also assigned under section Juliflorae in which comprises of around 235 species, characterize with having flowers in elongated spikes with numerous phyllodes, anatomize by longitudinal nerves (Hedge et al. 2013). Hedge et al. (2013) stated there are numerous studies that specify the growth rate of *Acacia mangium* tree could achieve the mean annual increment height of 5 m and diameter at breast height of 5 cm within the first 4–5 years. The tree will experience a rapid decline in growth after 7–8 years, depending on regional conditions, site and spacing. The tree height peaks at around 35 m with a diameter at breast height of 35 cm (Hedge et al. 2013).

Acacia mangium evergreen tree could thrive in most part of dense rain-forest, and tropical regions around the world. In regard to soil conditions, *Acacia mangium* thrive in low elevation and disturbed sites on well-drained acid soils of pH level of 4.5–6.5 (Hedge et al. 2013; Tanaka et al. 2015; Jang et al. 2004). A study conducted by Yamashita et al. (2008) goes in depth in their research regarding pH levels and soil inducing changes by *Acacia mangium* plantation in comparison with secondary forestry (Yamashita et al. 2008).

Climate wise, the distribution of *Acacia mangium* are within tropical warm and hot climate conditions, either humid or wet zones characterized by a short winter dry season and high total annual rainfall. In Bengkoka/Kudat region of Sabah, Malaysia, including the effects of seasonal conditions, the annual rainfall is over 2500 mm, hence it could be considered as an acceptable region for *Acacia mangium* tree to flourish (Hedge et al. 2013). A study conducted by Lee (2018), goes in depth regarding the development plantation and the future of *Acacia mangium* trees in both the Sabah and Sarawak region, in which share similar climate conditions (Lee 2018). The suitable temperature of the distribution area of *Acacia mangium* are within the mean maximum about 30–34 °C during the hottest month in the year and the mean minimum of about 15–22 °C.

Acacia mangium distribution area with the absolute minimum temperature falls below 0 °C is unsuitable, due to the possibility of developing frost (Hedge et al. 2013). The *Acacia mangium* area is not ideal if there are prolonged dry periods, as it will slow down the growth of the tree, as it under goes drought stress. Studies are

Fig. 13 *Acacia mangium* Willd. inflorescence in blooming stage (Hedge et al. 2013)

scarce regarding specifically *Acacia mangium* wood or trees of the acacia genus, hence most studies date back during the 80s and 90s.

Looking back from the past 5 years there is certainly a rise in the number of studies regarding type of *Acacia mangium* species, natural fibers, composite material, applicability, plantation and wood production (Hedge et al. 2013; Torres Vélez and Del Valle 2007; Krisnawati et al. 2011; N. R. Council 1983; Booth 1994). The anatomical features of *Acacia mangium* sapwood is white and sharply defined in comparison to the darker brown heartwood, where the wood has a fine texture and straight or interlocking grain. Hedge et al. (2013) reviewed numerous studies regarding *Acacia mangium*, where the researchers discussed not only in regards to the anatomical structure of *Acacia mangium* wood but also the overall cultivation from seed to a matured tree (Hedge et al. 2013) (Figs. 13 and 14).

9 *Acacia mangium* Willd. in the Malaysian Region

In 1967 the plantation of the *Acacia mangium* Willd. tree species was first introduced in Sabah, Malaysia. It was implemented into the region as a fire-break species (Tham et al. 1976). The prediction of a shortage of sawlogs for general timber was

Fig. 14 *Acacia mangium* Willd. planation in the Malaysian region (Lim et al. 2011)

forthcoming, hence a plantation program by the Forestry Department in Peninsular Malaysia, called the Compensatory Forest Plantation Program (CFFP) in 1982 (Lim et al. 2011). This program focused on the plantation-grown timbers within 3 regions of Malaysia, mainly the Peninsular Malaysia for *Acacia mangium*, Sarawak for engkabang jantong (*Shorea macrophylla*) and Sabah for teak (*Tectona grandis*) (Lim et al. 2011).

The plantation program established 188,000 ha for *Acacia mangium*, alongside *Gmelina arborea* and *Paraserianthes falcataria* as the main species of the forest. It was estimated that the yield of general utility sawlogs of 210 m^3/ha within 15 years. Specifically, regarding the plantation of *Acacia mangium* trees, a total of 51,768 ha were allocated for the plantation, mainly in the Peninsular region of Johor, Negeri Sembilan, Pahang and Selangor. For timber-based industries of Malaysia, commercial forest plantation could consider to be a step forward regarding the increase in production and meeting the government target of national export value earning of an estimated RM53 billion by the year 2020 (Lim et al. 2011).

The International Tropical Timber Organization (ITTO), launch a project on improving utilization and value adding of plantation timbers from sustainable sources in Malaysia. Majority of the reported studies are regarding properties of *Acacia mangium* planted in Peninsular Malaysia. One of the key sections of the study conducted was wood anatomy and quality of *Acacia mangium*. The evaluation conducted by the researcher on the properties of the plantation-grown *Acacia mangium* was from numerous forest reserve sites and of different age, mainly 20 and 16-year-old trees with average diameter of 37.9 and 32.5 cm respectively, with the height of 30 m and clear bole up to 6–12 m high.

The density of the *Acacia mangium* timber was reported to be increasing with age, and due to its fast growth-rate, the *Acacia mangium* timber tend to produce numerous number of knots, effecting the quality of the end-product.

The table shows the range of density of the *Acacia mangium*, at different ages and regions in Malaysia as reported by Lim et al. (2011), Lim and Gan (2000) (Table 5).

The acacia species in general demands the full exposure to sunlight in order to obtain good growth and development. There is possibility of growth stunting when there is lack of exposure to sunlight (Lim et al. 2011). On the other hand, some studies reported that the acacia species are susceptible to pest, pathogens and diseases potentially disrupt the health of the trees in the plantation (Lee 2018; Old et al. 2000; Tarigan et al. 2011). When pest, pathogens and diseases are affecting the plantation, quality and product yield are affected. In regard to the quality of the resulting timber, there are numerous factors such as sapwood and heartwood content, sunlight exposure, and plantation spacing of the tree and etc. (Hedge et al. 2013; Torres Vélez and Del Valle 2007; Tonini et al. 2018). The content of sapwood and heartwood varies according to the section of tree and age. It was noticeably reported that the density of the *Acacia mangium* tree timber increases as it aged. Due to the fast-growing nature of the *Acacia mangium* tree, the sapwood and heartwood content appears to be the opposite comparing to the density data. ITTO investigated the sapwood content of the *Acacia mangium* timber in order to evaluate the anatomical wood features of *Acacia mangium* timber and quality. Their investigation involves analyzing *Acacia mangium* timber from ages of 16 and 20 years old, where both samples were obtained from two different locations in the Malaysian region (Lim et al. 2011) (Fig. 15).

The figure illustrates the *Acacia mangium* log butt end cross section with the visible heartwood (dark color shade) and sapwood (light color shade).

Table 5 Density value of *Acacia mangium* Willd. at different ages and regions in Malaysia (Lim et al. 2011; Lim and Gan 2000)

Age (year)	Average basic density/range (kg m^3)	Regions in Malaysia
2	421	FRIM
5	290–500	Batu Arang, Selangor
6	340–500	Ulu Sedili, Johor
8	350–580	Ulu Sedili, Johor
9	389–535	Indonesia
12	570	Sabah
14	467–675	FRIM
16	522	Ulu Sedili, Johor
20	623	Kemasul, Pahang

Fig. 15 *Acacia mangium* Willd. log butt end cross section illustrating the heartwood and sapwood (Lim et al. 2011)

The concluding results of their investigation illustrated the percentages of sapwood content, basic density, and fiber length of the sampled *Acacia mangium* timber. The percentage of sapwood in 16-year-old *Acacia mangium* trees was discovered to be lower when compared to the 20-year-old *Acacia mangium* trees; 5.3–17.8% and 11.2–25.5% respectively. The basic density result supports the tabulated average basic density data statement regarding average basic density increases with age, where 522 ± 116.9 kg m^3 for the 16-year-old samples and 623 ± 102.6 kg m^3 for the 20-year-old samples. In terms of the average fiber lengths, ITTO results shown that the 16-year-old samples has an average fiber length of 954 μm and the 20-year-old samples has an average fiber length of 1048 μm. It could be correlated to the average basic density where aging and tree growth plays a factor to the average length of the fiber, older tree has longer fibers and higher density (Lim et al. 2011).

10 Sapwood and Heartwood (*Acacia mangium* Willd. and *Acacia melanoxylon* R.Br.)

In the wood production industry, sapwood and heartwood content of trees are investigated due to their individual aesthetics and characteristics. In general, sapwood could be distinguished by their light color shade compared to heartwood, this could be misleading due to wood coloring, as not all types of sapwood have a light color shade, and neither do the heartwood always has a darker color shade. Different tree species will have different percentages of sapwood and heartwood, as well as chemical composition, basic density, as well as mechanical properties. There are numerous factors related to the contribution towards the percentage of sapwood and heartwood in timbers, such as; growth rates, site conditions, species genetics, and age (Knapic et al. 2006). Numerous studies have investigated the growth of trees and timber as sawn materials in general to identify the characteristics and features of heartwood and sapwood (Blankenhorn 2001; Bradbury et al. 2011) Within the wood production industry, heartwood content is considered to be more desirable as compared to sapwood content, due to the sapwood being susceptible to fungi, has a higher moisture content and more susceptible to warping or shrinking, hence it is considered to be a quality variable (Tenorio et al. 2012). Apart from ITTO investigation on *Acacia mangium* sapwood and heartwood percentages. An investigate the heartwood and sapwood variation but with a different acacia tree species which is *Acacia melanoxylon* R.Br., commonly known as Australian blackwood (Knapic et al. 2006). The Australian blackwood samples were obtained by the researchers were from 4 different sites located in northern and southern Portugal (Figs. 16 and 17).

In this investigation, the Australian blackwood the age of the tree was not measured as compared to the ITTO project on *Acacia mangium* trees. However, the height of the tree used as timber samples were similar, where the shortest height of the tree used was 28.6 m and the highest is 30.4 m (Knapic et al. 2006). In their concluding results, their investigation only concerned the percentage of heartwood in the Australian blackwood samples at varied percentage of the tree height. From the overall height of the sampled tree, the highest content of heartwood is 61.3 ± 7.8% and the lowest content is 59.8 ± 9.7%. Comparing the *Acacia mangium* and the

Fig. 16 Blooming flower of the *Acacia melanoxylon* R.Br. tree (Tropical Plants Database 2019)

Fig. 17 Forest thriving with *Acacia melanoxylon* R.Br. trees (Invasive.org. 2018)

Australian blackwood content of heartwood content of the overall tree, it could be considered that the *Acacia mangium* has a higher content of heartwood compare to the Australian blackwood tree (Knapic et al. 2006; Igartúa et al. 2017). Some studies report the influences of heartwood and sapwood content in relation to the potential applications of the general acacia species (Lourenço et al. 2008; Baqui et al. 1984; Chang et al. 2017; Tham and Liew 2012; Lo and Liew 2011).

The sapwood percentage of the *Acacia mangium* at highest percentage is 25.5% of the overall tree height, as according to the ITTO project (Lim et al. 2011; Knapic et al. 2006). Hence, *Acacia mangium* tree plantation would be beneficial if wood industries are demanding an acacia species timber with relatively high heartwood content. On the other hand, the researchers also noted that the acacia species is considered as an invasive species and specifically difficult to control in the forest of Portugal, hence wood industries should take initiative towards the application of using acacia trees as quality timber products (Knapic et al. 2006). Assisting the surrounding flora and controlling the invasive species from overtaking the forest wellbeing.

11 Summary

In summary, there is a plethora of acacia species in tropical regions of the world, where majority are considered as an invasive species towards other flora in the region. Wood industries could consider the acacia species as potential resource for timber products as it has high percentage of heartwood desirable for wood working applications, and as a method to control the invading species. Plantation projects such as what ITTO has reported of the acacia species could serve as potential main resource for wood industries, while some may identify specific concentration area in different regions to control the invasive species.

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Acacia Wood and Its Surface Treatment for High Strength Bio-composites

E. Jayamani, M. Zain, Md Rezaur Rahman and M. K. B. Bakri

Abstract Till date, the core issue amongst many ecological problems that we face has been about nature and its various pollutions. Wood and natural fiber were known to be the very first materials used to combat noise pollution due to its miraculous properties of high strength and high sound absorption. It was employed as the initial material to reduce the high levels of noise in the harsh environment. Gradually, wood, natural fiber and its waste have become abundant. Moving on, agricultural lignocellulosic materials are also seen as environmentally friendly, reproducible and sustainable fibers. To counter this problem, researchers headed towards the use of synthetic materials that could reduce this problem. It is known that natural fiber and wood composite materials are analyzed to be weak in mechanical properties when compared to synthetic polymer composites, but the sustainability factor insisted the researchers to serve the cause of improving the mechanical properties of natural fiber composite materials. Various methods have now been established to improve and analyze the surface characteristics of wood and natural fibers and hence the mechanical properties of natural fiber composite materials. In this study, a thorough literature review is conducted on the chemical and physical treatment of wood and lignocellulosic residues and their characterization.

Keywords Acacia wood · Natural fibers · Surface treatments · Composites

1 Introduction

In the present modern era, a prominent portion of composite materials engineered materials take-up and ranging from day to day life products to complex and bigger applications (Verma et al. 2012). Composite materials are known as weight-

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E. Jayamani (✉) · M. Zain
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia
e-mail: ejayamani@swinburne.edu.my

Md R. Rahman · M. K. B. Bakri
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia

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saving materials. The composite industry has identified that numerous innovative experimental techniques through research and development produces some of these costs-efficient materials. However, the improvement in these composite materials in terms of cost cannot be brought through manufacturing techniques alone. There must be a programmed integrated effort to bring together various aspects such as material, process, design, quality assurance, manufacturing, tooling, and management for composites to become competitive and economic compared with metals or other materials. Due to the sheer size of the transportation industry, the industry has begun to distinguish that the commercial applications of composites promise to offer much larger business opportunities than the aerospace sector. There has been a clear shift of applications that were first centered onto the aircraft assemblage to other prospective and potentially sound commercial uses in the recent years.

The penetration of these advanced materials has witnessed a firm extension in the uses and volume, which increasingly enabled by the introduction of newer polymer resin matrix materials and high-performance reinforcement, such as fiberglass, carbon, aramid, wood and other natural fibers. Due to this, there has also been a decrease in the cost due to increased volume. The usage of composites instead of metals has largely reduced the cost and weight factors, as noticed in some applications. The common examples are engines cascades, replacements for weld metallic parts, ducts, curved fairing and fillets, cylinders, tubes, blade containment bands, etc. On the other hand, more seismic resistant structures and the requirement of a composite of its lighter construction materials has placed high importance on the use of new and advanced materials. It not only decreases the weight, but also improves the required mechanical properties such as shock and vibration absorption through tailored microstructures. Ecological complications have made us ponder over the use of more natural substances, which includes recyclability and environmental safety are both assured this way. Manufacturers must take this factor into account that every introduction of materials comes up with full advantage of environment friendly nature. Design for recycling is a much larger perspective; eco-design has been incorporated as a key philosophy being applied into the manufacture of every new material and product.

2 Wood, Natural Fiber and Its Polymer Interface

Saheb and Jog (1999) mentioned that the principle of the reinforcement in a composite material depends on the mechanical properties of the neat resin system. Unique properties possess by different fibers of a composite eventually affecting the overall properties and efficiency of the composite material. To assure proper and neat handling, these fibers must be assembled into a sheet like conformation, known as fabric.

Saxena et al. (2011) stated that the interface is a bounding surface or zone where all these discontinuities occur and if the individual components may fail to depict, if any of its characteristics are in complete isolation, either its physical, mechanical, chemical or any other nature. The matrix of the composites must cover or "wet" the fiber. To improve wet ability, enhancer or coupling agents are known to be frequently used. Well "wetted" fibers increase the interface surface area. The interface must be able to pass through the applied load from matrix to fibers. These steps are essential to increase the overall attainment of the desirable properties. This would demonstrate strong adhesion between fibers and matrix. Failure at the interface (called debonding) may or may not be desirable. Coupling agents are molecules possessing two functions. The first is to react with OH groups of cellulose and the second is to react with functional groups of the matrix. The choice for a coupling agent that can combine both the properties of strength and toughness to a considerable satisfying degree is an important aspect to keep in mind, while manufacturing of a composite material.

2.1 Classification of Natural Fiber

The fibers are generally categorized into two classes, man-made or synthetic and naturally existing fiber. Natural fibers include those made from plant, animal and mineral sources. Like pieces of threads, fibers make up the class of hair like materials much like continuous filaments or discrete elongated fragments (Saheb and Jog 1999). They can be converted into bigger shapes like filaments, threads and ropes (Saheb and Jog 1999). Most of them are compromised in major components of the composite materials. Most of these fibers were transformed into mat sheets. Animal fibers are generally based on proteins, such as mohair, wool, silk, alpaca, angora (Saheb and Jog 1999). Animal hair (wool or hair) is usually taken from animals or hairy mammals, e.g. sheep's wool, goat hair, alpaca hair, horse hair etc. Silk fiber is collected from dried saliva of bugs or insects during the grounding of cocoons, such as silk from silkworms. While, avian fibers are from birds, e.g. feathers and feather fiber. Mineral fibers are naturally occurring fiber or slightly modified fiber procured from minerals (Kindo 2010). These were categorized into the following categories: asbestos, as the only naturally occurring mineral fiber. Variations are serpentine and amphiboles, anthophyllite. Ceramic fibers such as glass fibers (glass, wood and quartz), aluminum oxide, silicon carbide, and boron carbide. Metal fibers, such as aluminum fibers.

Plant fibers are mainly composed of cellulose, for examples include flax, ramie, sisal, cotton, jute, and hemp (Taj et al. 2007). In the manufacturing of paper and cloth, cellulose fibers also have an application. This fiber can be further categorized into following: seed fibers, which is collected from the seed and seed case e.g. cotton and kapok. Leaf fibers, which collected from the leaves e.g. sisal and agave. Skin fibers are collected from the skin or bast surrounding the stem of their respective plant. These fibers have higher tensile strength than other fibers. Therefore, these fibers are used for durable yarn, fabric, packaging, and paper. Some examples are hemp, flax, jute, banana, and soybean. Fruit fibers are collected from the fruit of the plant, e.g. coconut (coir) fiber. Stalk fibers are the stalks of the plant, e.g. straws of wheat, rice, barley, and other crops including bamboo and grass. Tree wood is also such a fiber.

2.2 Polymer Matrix

A very large number of polymeric materials, such as both thermosetting and thermoplastic are used as matrix materials for the composites (Taj et al. 2007). In general, the polymer matrices are selected based on adhesive strength, fatigue resistance, heat resistance, chemical and moisture resistance etc. The resin must have mechanical strength proportionate to that of the reinforcement. It must be convenient to use in the fabrication process selected and have a way to go side by side with the service conditions. Apart from these properties, the resin matrix must be capable of wetting and deepening into the bundles of fibers which provide the reinforcement, replacing the dead air spaces therein and offering those physical characteristics capable of bolstering the performance of fibers. Thermosetting resins, such as polyurethane, phenolic, epoxy, polyester, etc. are commonly used today in natural fiber composites, in which composites requiring higher performance applications. They provided plenty of mechanical properties, such as stiffness and strength, at moderately low-price levels. Replacing fibers with natural materials is the first step in considering the paramount importance of ecological aspects. Lowering down the emission of greenhouse effect, causing gases such as CO_2 into the atmosphere and an increasing awareness of the finiteness of fossil energy resources are helpful in developing new materials that are entirely based on renewable resources.

2.2.1 Synthetic Fiber Reinforced Polymer

Common fiber reinforced composites are composed of fibers and a matrix (Saheb and Jog 1999). Fibers act as a reinforcement and the main source of strength, while matrix bonded all the fibers together in shape and transferred the stresses between the reinforcing fibers. Most of the fibers act as a carrier that carries the loads along their longitudinal directions. Sometimes, filler might be added to smooth the manufacturing process, inculcate special properties to the composites, and/or reduce the product cost. Common fiber reinforcing agents includes beryllium, beryllium asbestos, carbon/graphite fibers, carbide, beryllium oxide, aluminum oxide, glass fibers, polyamide molybdenum, synthetic natural fibers, etc. Similarly, common matrix materials include polyester, polyurethane, epoxy, phenolic, vinyl ester, polyetheretherketone (PEEK), etc.

2.2.2 Mineral Particle Reinforced Polymer

Ceramics and glass particles are examples of particle used for reinforcing, metal particles such as aluminum and amorphous materials are also one of its, including polymers and carbon black (Verma et al. 2012). They can easily increase the modulus of the matrix, as the particles are usually used to decrease the ductility of the matrix. These types of particles are also used to reduce the cost of the composites, either in term of reinforcements and matrices, which are commonly cheap, and the material can easily be processed. Some of these constructive properties of ceramics and glasses, may cause high melting temperature, high strength, stiffness, low density, wear resistance, and corrosion resistance. Many ceramics are good electrical and thermal insulators. Some ceramics have extraordinary properties, which posed as magnetic materials; some are piezoelectric materials; and a few special ceramics are even superconductors at very low temperatures.

2.2.3 Wood and Natural Fiber Reinforced Polymer

The interest in wood and natural fiber-reinforced polymer composite materials are rapidly growing both in industrial applications and fundamental research (Saxena et al. 2011). They are renewable, completely or partially recyclable, cheap, and biodegradable. Plants, such as cotton, hemp, jute, kenaf, flax, pineapple, sisal, ramie, bamboo, banana, etc., as well as wood, which are used from time immemorial as a source of lingo cellulosic fibers, are more and more often applied as the reinforcement of composites. Their availability, reliability, renewability, low density, and price as well as their satisfactory mechanical properties, make them an attractive ecological alternative to glass, carbon and man-made fibers used for the manufacturing of composites. The wood and/or natural fiber-containing composites are more environmentally friendly, and are used in transportation (automobiles, railway coaches, aerospace), military applications, building and construction industries (ceiling paneling, partition boards), packaging, consumer products, etc.

3 Treatment

Several treatments of wood fibers may be considered. As per their methodological aspects and the agents used, it can be classified into two broad categories: physical treatment and treatment.

3.1 Physical Treatments

3.1.1 Plasma Grafting

Physical treatments have a way of changing the structure and surface properties of the fiber, as shown in the result of the mechanical bonding with the matrix (Zafeiropoulos 2011). A very intriguing and an environmentally reliable surface treatment concerning the use of atmospheric air pressure plasma (AAPP) was applied to various lignocellulosic fibers, such as abaca, flax, hemp and sisal via treatments narrowed down to a few minutes. The treated surface's wettability measure was determined using capillary rise technique and by applying the zeta potential measurements, whereas the changes in the surface chemistry were also characterized. Fibers' swelling behavior that comes under the umbrella term of bulk properties of the fibers was also looked upon. Even for increased and sustained timings, the surface energy of the lignocellulosic fibers was seen to have remained practically constant with one exception being abaca fibers. The parameter decreased with increasing AAPP treatment time over abaca fibers.

3.1.2 Other Techniques

Zafeiropoulos (2011) stated that by lending the super-hydrophobic properties, as that of an original method over the paper surfaces through alkyl ketene dimer in a crystalline form, it shown significant improvement. Through that, there are three diverse techniques can be used, such as (i) air blasting with cryo-ground microparticles; (ii) crystallizing from organic solvents; and (iii) spraying with rapid expansion of supercritical solutions. All these three techniques gave satisfactory results; however, the third technique came out to be producing the most reliable and sufficiently effective results out of the three as far as the surface methodology is concerned. There has been a clear insight regarding the measurements of wetting, but no such observation about the long-termed stability of the physical coatings was established. The very significant of applications appeared to be in the industrial implementation, where the usage and the potential of this study imply an application in a continuous manner without having to use the solvent.

3.2 Chemical Treatments

3.2.1 Alkaline Treatment

George et al. (2004) reports that the alkaline treatment is the most widely chemical methods exploited over the natural fibers. The impacts of alkaline treatment bring surface roughness by disrupting hydrogen bonding in the network structure. This treatment does two things: it removes the wax, lignin and oils coating the external surface of the fiber wall to some extent and depolymerizes cellulose. Because of which the short length crystallites are brought to exposure.

3.2.2 Silane Treatment

George et al. (2004) reports that silane has the chemical formula of SiH_4. These chemical compounds stabilize the composite material by being coupling agents to let glass fibers adhere to a polymer matrix. Silane coupling agents have been seen to reduce the number of cellulose hydroxyl groups in the fiber-matrix interface.

The hydrolysable alkoxy group leads to the formation of silanols in the presence of moisture. Stable covalent bonds are established with the cell wall whenever Silanol reacts with the hydroxyl group in the fiber that are then chemisorbed onto the fiber surface. This strong covalent bonding between the matrix and the fiber results into a formidable cross-linked network of the hydrocarbon chains that further pins down the swelling of the fiber, a considerable result achieved through this silane treatment.

3.2.3 Acetylation of Natural Fibers

Li et al. (2007) reported acetylation processes is to bring an acetyl functional group (CH_3COO^-) into an organic compound is a reaction. Plasticization of cellulosic fibers were achieved through esterification methods, which also known as the acetylation of natural fibers. Before the fiber is employed to usage, the by-product acetic acid (CH_3COOH) of the reaction is necessary to be removed from the lignocellulosic material. Chemically modified properties of the polymers through hydrophobic nature is obtained, whenever acetic anhydride ($CH_3–C(=O)–O–C(=O)–CH_3$) replaces the polymer's hydroxyl groups of the cell wall with acetyl groups.

3.2.4 Benzoylation Treatment

Li et al. (2007) reported that benzoylation are one of the methods known for organic synthesis. The fiber treatment used an important agent like benzoyl chloride. The reinforced interaction with the hydrophobic PS matrix together with the downfall in the hydrophilic nature of the fiber are granted by the presence of the benzoyl group ($C_6H_5C{=}O$) in the benzoyl chloride.

3.2.5 Acrylation and Acrylonitrile Grafting

Mohanty et al. (2001) reported that acrylation occurs by the presence of free radicals of the cellulose molecule. Cellulose was employed to generate radicals within the chain scission during the treatment, with high energy radiation. Glass fibers are also modified by the graft polymerization of the acrylic acid ($CH_2{=}CHCOOH$). The treatment has often proved to be effective for the enhancement of natural fibers.

3.2.6 Malleated Coupling Gents

Keener et al. (2004) reported that to strengthen the composite materials containing fillers and fiber reinforcements, malleated coupling agents are widely used. In the method of applying chemical treatments, it was to keep the usage of maleic anhydride, but for PP matrix, it was incorporated into the studies to have better interfacial bonding and mechanical properties in composites. The PP matrix chain is a great way to produce maleic anhydride grafted polypropylene (MAPP) and allows the maleic anhydride to be cohesive. Strong covalent bonds across the surface were exorbitant and were achieved in treating the cellulose fibers with hot MAPP.

3.2.7 Permanganate Treatment

Permanganate is a compound containing permanganate group MnO_4^- (Kalia et al. 2009). Through MnO_3^- ion formation, cellulose radical is formed when treatment is carried out using Permanganate. By initiating the graft polymerization, highly reactive ions of Mn^{3+} are generated. The majority of the permanganate treatments is equipped with the usage of potassium permanganate ($KMnO_4$) solutions.

3.2.8 Peroxide Treatment

Li et al. (2007) reported that peroxide is a specific functional group or a molecule with the functional group ROOR containing the divalent ion O–O in organic chemistry. Decomposition of the organic peroxides into the free radicals form of RO–; RO– then reacts with the hydrogen group of the matrix and cellulose fibers benzoyl peroxide (BP, $((C_6H_5CO)_2O_2)$ and dicumyl peroxide (DCP, $(C_6H_5C(CH_3)_2O)_2)$, which are chemicals in the organic peroxide family that are used in natural fiber surface modifications.

3.2.9 Isocyanate Treatment

Mohanty et al. (2001) reported that the compound containing an isocyanate functional group –N=C=O is called an isocyanate and is highly vulnerable to react with the hydroxyl groups of cellulose and lignin in the fibers. It usually acts as a coupling agent in the utilized of fiber-reinforcement of composite. The chemical treatments have resulted increased in highly producing interface adhesion between the matrix and fiber and decrease the overall water absorption of the fiber. Therefore, the chemical treatments were taken as a great source of method modifying the properties of the natural fibers.

4 Characterization of Fiber Surface Treatments

There have been plenty of ways to characterize the fiber surface based on the properties and to analyses the fiber surface. Many researchers have discussed them in detail.

4.1 Fourier Transform Infrared Spectroscopy

The most widely incorporated studies have been through Fourier transform infrared (FTIR) spectroscopy. Zafeiropoulos (2011) reported that most of it is done based on the surface techniques incorporated with it to show the correlation between both techniques. Almost all materials have been seen to absorb the infrared radiation and the frequency intervals, which the absorption bands appear to be associated with the vibrational modes of specific functional groups. Most of the interpretation and representation of the values obtained by scientists are collected over the large durations of their continuous research, which is as large bodies of infrared data presented in the tabulated form and charts. Most of this information is now accessible on the internet and through software libraries. The dipole moment of the molecule must change to keep the vibration motion to be IR active, the higher the magnitude of the change is, the higher the intensity of the band will be achieved. As the functional group is usually associated with more than one absorption band (more than one vibrational mode) and different functional groups may absorb in overlapping regions.

4.2 Raman Spectroscopy

Colthup et al. (1990) reported that Raman spectroscopy is also based on the vibrational motion of functional groups. It detected the changes in the polarizability of the molecule. The method is however are the least used these days. This polarizability decreases with increasing electron density, increasing bond strength and decreasing bond length. There has been an utilization of the characteristic in determining extension ratios in fibers/microfibers embedded in different composites.

4.3 Single-Fiber Fragmentation Test

Tripathi and Jones (1998) reported that the single-fiber fragmentation test (SFFT) method was originally used with metals by Kelly and Tyson. They observed a multiple-fiber fracturing phenomenon in a system, consisting of a low concentration of brittle tungsten fibers embedded in a copper matrix. The main assumptions made by Kelly and Tyson were that the matrix yields plastically, the fiber-matrix adhesion is perfect and the shear stress along the fiber is constant and equal to the shear yield strength of the matrix. A single fragmentation test to measure the interfacial shear strength was available for those composite systems, which the ultimate elongation of the matrix is higher than the fiber elongation. Most of the test proposed that the strain at matrix fracture must be at least three times higher than that of the fiber. The method involved completely embedding a single fiber along the centerline of a relatively large dog-bone shaped specimen of matrix material. The specimen was then pulled in tension uniaxially along the fiber axis. Stress was transferred to the fiber by the matrix. As the strain was increased, the fiber broke. The matrix restrained the fiber fragments for returning to their unstressed dimension when the fiber first broke.

4.4 Fiber Pull-Out Test

Colthup et al. (1990) reported that although the fiber pull-out test is popular due to its conceptual simplicity, but there are some difficulties may present. A fiber was embedded in a very thin film or bead of the polymer and the specimen was gripped in a tensile testing machine for testing. The fiber was pulled out of the polymer and the force required to remove the fiber was measured. The adhesive strength was calculated by dividing the measured maximum load on the area of contact of the fiber with the polymer. The area of contact was measured using optical microscopy. However, most of the fibers are cylindrical with a smooth surface, which may not be entirely validated, and it therefore influences the accuracy of calculation because of difficulties in measuring the exact embedded surface area for each fiber.

4.5 Micro-debonding Test

Zafeiropoulos (2011) reported that the modified version of the pull-out test is the micro-debonding test. The test was established for the characterization of single fiber-matrix interface, which is to remove any meniscus effect. In this test, a small drop of the matrix was deposited onto the fiber at some point. The fiber with the microbead was mounted in a micro-device and then the fiber is pulled out. The test has been widely used to evaluate τ for both thermoset and thermoplastic matrix composite, due to simplified sample preparation. Despite the ease of performing this test, there are several concerns that must be taken into consideration during the performance, such as stress concentrations during specimen loading, non-uniform shear stress distribution along the fiber-matrix interface, the geometry of the polymer drops, and the effect of the strain rate. All these factors significantly affect the test results and their scatter.

5 Effect of Different Chemical Treatments on the Different Fibers Performance

5.1 Wheat Straw

Durot et al. (2003) treated the wheat straw with two alkali's, NaOH and K_2CO_3. Extractive wheat straw (EFW; 50 mg) was incubated for 24 h and stirring at room temperature at 20–25 °C in alkaline solution (10 mL). Two reaction media were used: (i) potassium carbonate, mol/L, pH = 10 (adjusted with hydrochloric acid); (ii) sodium hydroxide, 0.1 mol/L, which carefully titrated to pH = 10 with dilute HCl solution. At the end of the reaction period, the straw was separated from the alkaline medium through filtration.

It is reported that a considerable fraction of lignin was solubilized in the reaction media, which was expected around 32.6 and 16.5% delignification for K_2CO_3 and NaOH treated samples, respectively. The structures were analyzed by thio-acidolysis. The CO_3–Lig contained three times fewer ether-linked monomers than the OH–Lig and ref–Lig. The wheat straw cell wall undergone structural modifications, which promoted by the treatment of potassium carbonate and sodium hydroxide, that lead to the revealing of some lignin structures, as the study has depicted. It was observed that after the treatment, K_2CO_3 solubilized a greater amount of lignin that contained more condensed structures than the NaOH at pH 10.

5.2 Banana Fiber

Ahad et al. (2009) treated banana fiber and coconut husk with NaOH. To obtained long straight fibers, banana fibers were separated by water retting and scraping. The fibers were then washed with water and dried under the sunlight. The fibers were engrossed in NaOH solution with different concentrations of 5, 10 and 15% for 3 h at room temperature. After treatment, the fibers were washed with water again. SEM was employed to observe the surface morphology of untreated and treated fibers. Results revealed that impurities from the natural fibers were removed from the banana fiber upon treatment with the alkaline (NaOH) treatment.

5.3 Sisal Fiber

Mwaikambo and Ansell (2002) used NaOH to treat the sisal fibers. Mwaikambo and Ansell (2002) sodden the fibers in beakers that contained different NaOH concentrations and were kept in a water bath and guarded at 20 °C for 48 h. The fibers were then removed, washed with distilled water containing 1% acetic acid to neutralize excess sodium hydroxide. It was analyzed that sisal fibers possessed shorter length fiber, which are joined together to end, and they were discontinuous. Figure 1a, b shows the SEM results of treated and untreated sisal fibers.

Li et al. (2011) used MPS-g-PLA (Polylactidegraft-γ methacryloxypropyltrimethoxysilane) and PLA-co-PGMA (Polylactide-co-glycidyl methacrylate) to treat sisal fiber. The MPS-g-PLA treatment of sisal fiber was carried out by immersing the fibers in the dioxane solution of MPS-g-PLA at the concentration of 1 wt% for 48 h. Sisal fibers were taken out of the solution and dried for 2 days at room temperature. For PLA-co-PGMA treatment, sisal fibers were absorbed in the dioxane solution of PLA-co-PGMA at the concentration of 1 wt% for 48 h. After drying it at room temperature, the products were soxhlet extracted with THF for 24 h and before being dried again. The treated and untreated fibers were fabricated with PLA, while the flexural and impact strength were tested, results of which are shown in Figs. 2 and 3.

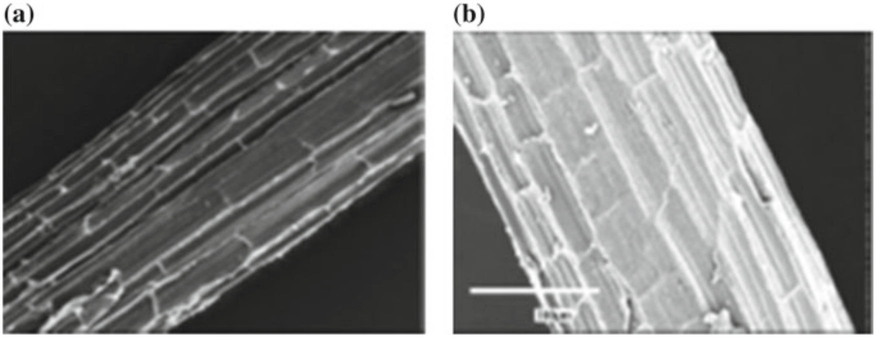


Fig. 1 **a** Treated sisal fiber, and **b** untreated sisal fiber

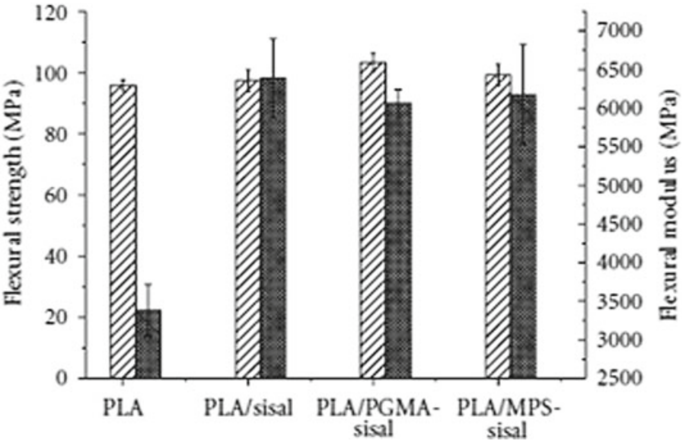


Fig. 2 Flexural properties of 30 wt% surface-treated sisal fibers compared to untreated sisal fibers reinforced composites

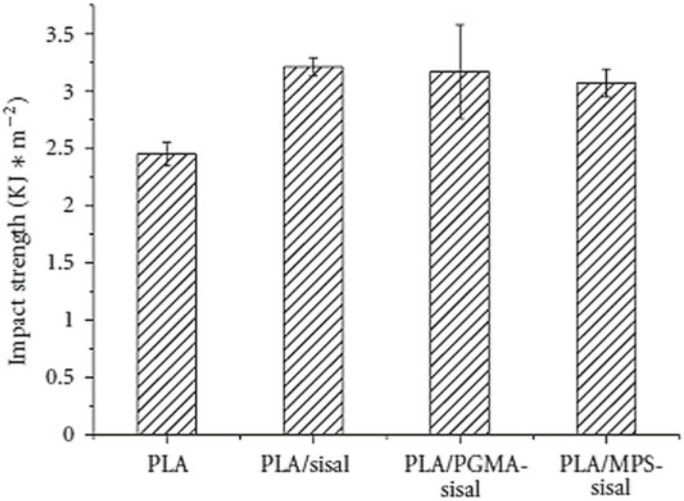


Fig. 3 Notched Izod impact strength of sisal fibers reinforced PLA composites

5.4 *Coconut Husk Fiber*

Ahad et al. (2009) treated coconut husk fiber with NaOH. Coconut husk fibers were separated by water retting and scraping. The fibers were then subjected by washing it with water and dried under the sunlight. The fibers were soaked in NaOH solution with different concentrations of 5, 10 and 15% for 3 h at the room temperature. After treatment, the fibers were washed under running water and dried at room temperature for two days. Figure 4 shows the SEM images of coconut.

Above are the results of SEM for the treated coconut fiber. It was analyzed that the specimen treated with 15% NaOH had smother surface than 5% NaOH treated specimen. It was analyzed that the treated fiber had a better surface roughness, which is a good stance in creating better interlocking between the fiber and the matrix.

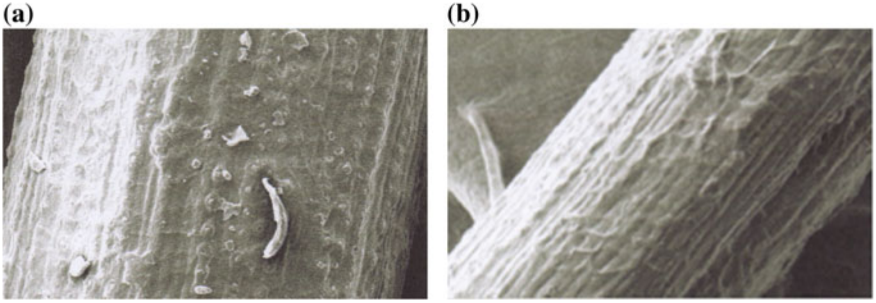


Fig. 4 Coconut fiber after treatment at 300× magnification **a** 5% NaOH, and **b** 15% NaOH

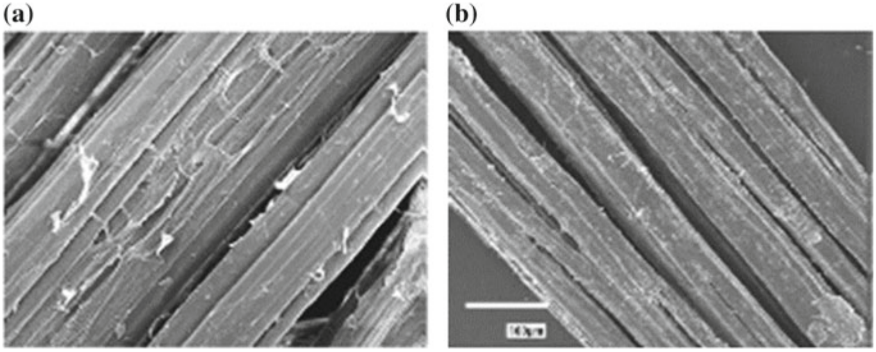


Fig. 5 **a** Untreated hemp fiber, and **b** treated hemp fiber

5.5 Hemp Fiber

Mwaikambo and Ansell (2002) treated hemp fiber with NaOH and acetic anhydride. Fibers were soaked and washed in cold distilled water for 48 h at 20 ± 2 °C to eliminate any surface impurities. Fibers were soaked in beakers containing different NaOH concentrations and were kept in a water bath guarded at 20 °C for 48 h. The fibers were then removed, washed with distilled water containing 1% acetic acid to neutralize the excess sodium hydroxide before carefully rinsed with distilled water. Fibers were then treated with acetic anhydride containing concentrated H_2SO_4 as a catalyst for 5 min. Fibers were then washed with distilled water and dried. SEM was employed to evaluate the surface characteristics of treated hemp fiber. It was analyzed that the hemp fibers were continuous in their outlook. Treated hemp fiber looked cleaner and refined along with the fiber bundles set apart in a separated fashion, with a highly serrated surface. SEM results are shown in Fig. 5.

Bismark et al. (2004) treated hemp fiber with NaOH and MAH-graft PP. It was reported that the fibers were boiled in 2–10% sodium hydroxide solutions. The fibers were soxhlet-extracted with toluene for 24 h and then dried at 60 °C under vacuum (1 mbar) for 24 h. The purified fibers were then immersed in a solution of (3 and 5%) MAH-graft PP (concentration: 5 wt% on the fibers) in hot (100 °C) toluene for 5 min. After grafting the fibers, it was again soxhlet-extracted for 48 h, to remove all components, which were not covalently attached to the fibers. Finally, the fibers were dried again at 60 °C under vacuum (1 mbar) for 24 h. IR measurements gave evidence for covalent bonding of the modified PP to the natural fibers.

5.6 Rice Straw

Kamel (2004) treated rice straw with NaOH and coupling agent bagasse lignin. The rice straw was subjected to treatment with various concentrations of NaOH for 1 h at different temperatures. The fibers were then washed with water, while having slight amounts of acetic acid in it to remove the ample amounts of NaOH being caught up between the fibers before being dried again. Eight samples were prepared, which were treated at different concentrations at different temperatures. They were fabricated with PVC. Several tests were carried out to analyzes the effect of chemical treatment of rice straw results, which are shown in Figs. 6 and 7. It was clear from Kamel's work that differently treated rice straw resulted in composite with different mechanical properties.

5.7 Henequen Fiber

Lee et al. (2008) treated the straight henequen fibers with NaOH and treated them physically by using ultrasonic frequency. Fibers were soaked in water at 30 °C for 2 h. Henequen bundles were also treated with 1 wt% and 6 wt% NaOH aqueous solutions for 10 min and 60 min, respectively. The ultrasonic frequency applied was 40 kHz. The fiber bundles were washed with distilled water for several minutes after alkalization, until pH 7 was achieved. The treated fibers were fabricated with polypropylene and then tested for various mechanical properties. Results are shown in Figs. 8 and 9.

From the results, it shows that the soaking (static method) and ultrasonic (dynamic method) treatments with tap water and sodium hydroxide at different concentrations and treatment times, significantly influenced the interfacial, flexural and dynamic mechanical properties of henequen/polypropylene bio-composites. Compared with the water treatment, the alkaline treatment was more effective in improving the interfacial and mechanical properties of randomly oriented, chopped henequen/PP bio-composites.

5.8 Flax Fiber

Bismark et al. (2004) analyzed the green flax, which were treated with NaOH and MAH-graft PP. It was reported that by boiling the fibers in 2–10% sodium hydroxide solutions, they were purified. The fibers were soxhlet-extracted with toluene for 24 h and then dried at 60 °C under vacuum (1 mbar) for 24 h. The purified fibers were then immersed in a solution of (3 and 5%) MAH-graft PP (concentration: 5 wt% of the fibers) in hot (100 °C) toluene for 5 min. After grafting the fibers, it was again Soxhlet-extracted for 48 h to remove all other components, which were not covalently attached to the fibers. Finally, the fibers were dry again at 60 °C under vacuum (1 mbar) for 24 h. IR measurements gave evidence for covalent bonding of the modified PP to the natural fibers. IR measurements on MAH-graft PP cellulose- and flax fibers gave evidence for covalent bonding of the modified PP to the natural fibers. The measured weight gain after grafting 3% MAH-graft PP on a flax sample

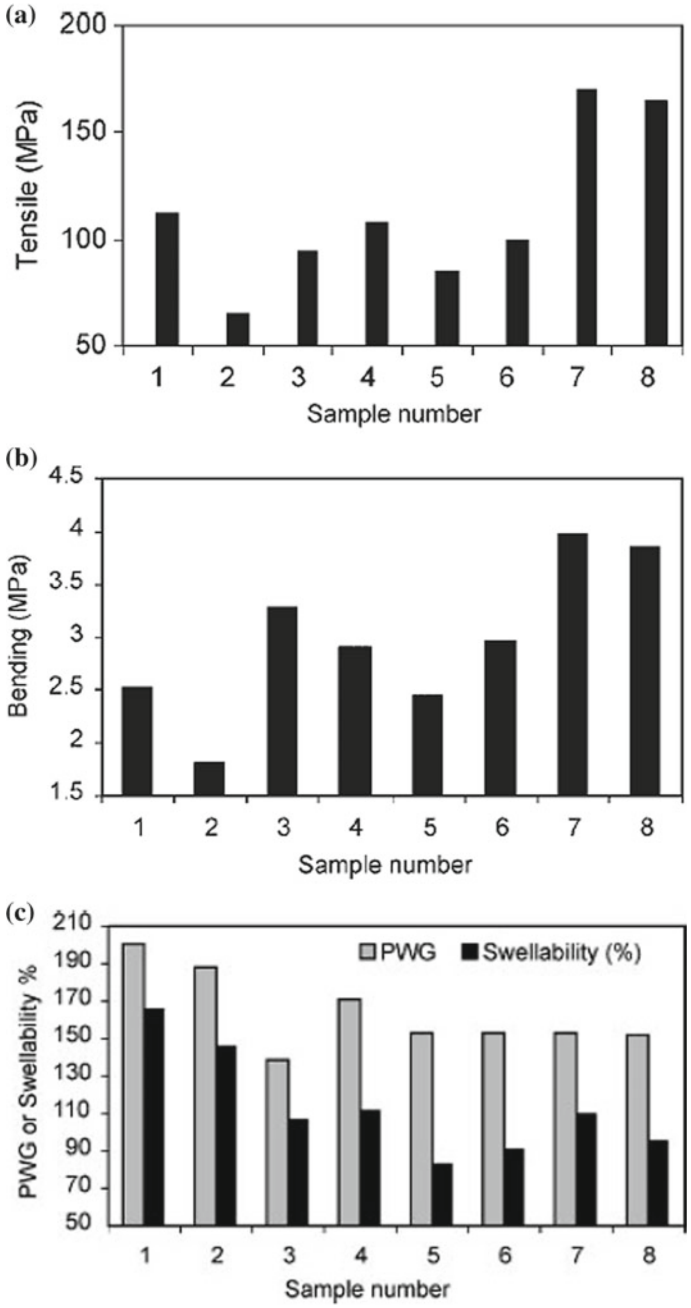


Fig. 6 Effect of pre-treatments of rice straw on **a** tensile strength, **b** bending strength, and **c** PWG or swellability % of the composites

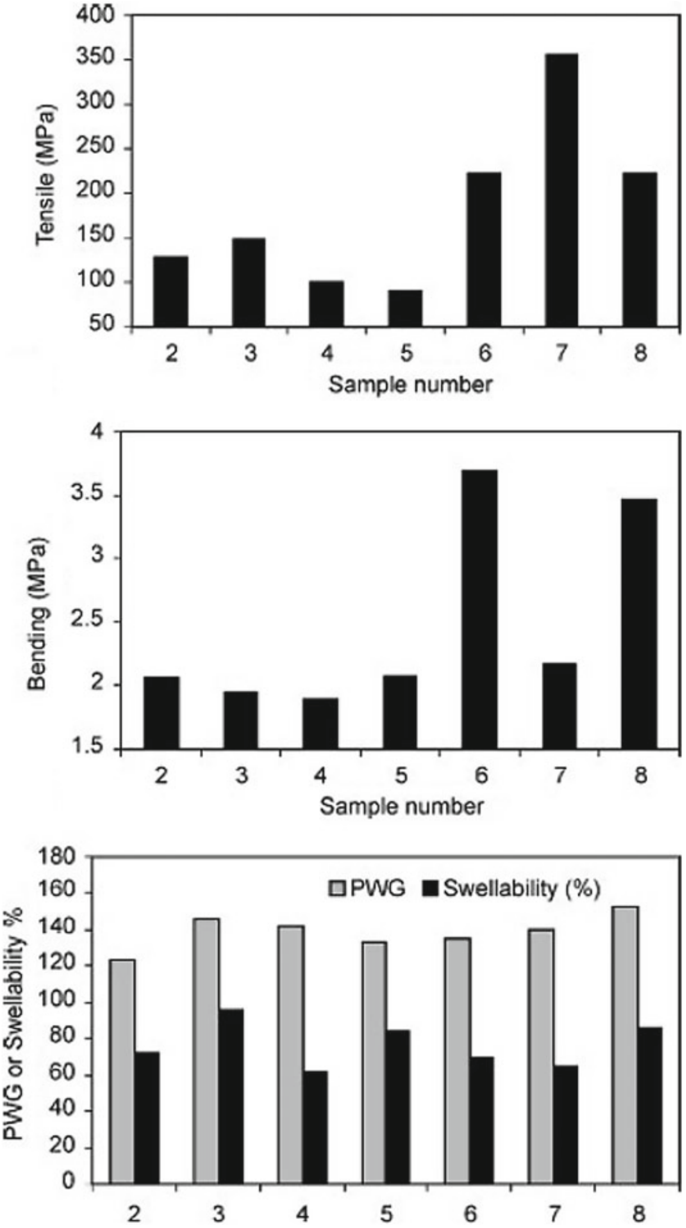


Fig. 7 Effect of bagasse lignin (3%) on **a** tensile strength, **b** bending strength, and **c** PWG and swellability % of composites made from different pre-treatments of rice straw

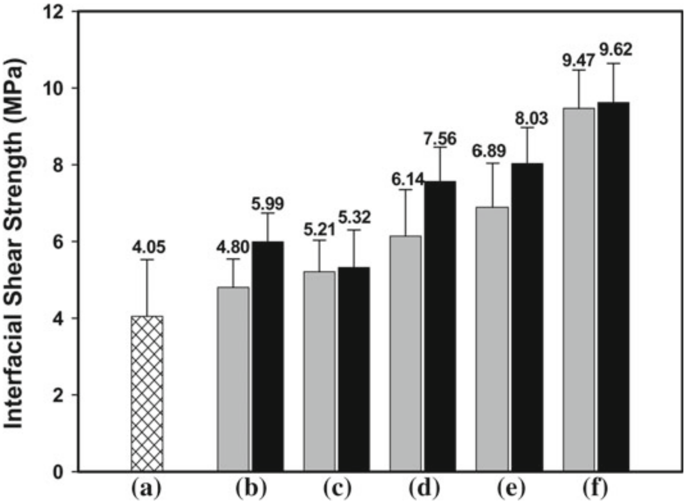


Fig. 8 Interfacial shear strengths of **a** henequen/PP bio-composites, **b** untreated and treated by static (gray) and dynamic (black) methods with tap water, **c** NaOH 1 wt%, 10 min, **d** 1 wt%, 60 min, **e** 6 wt%, 10 min, and **f** 6 wt%, 60 min

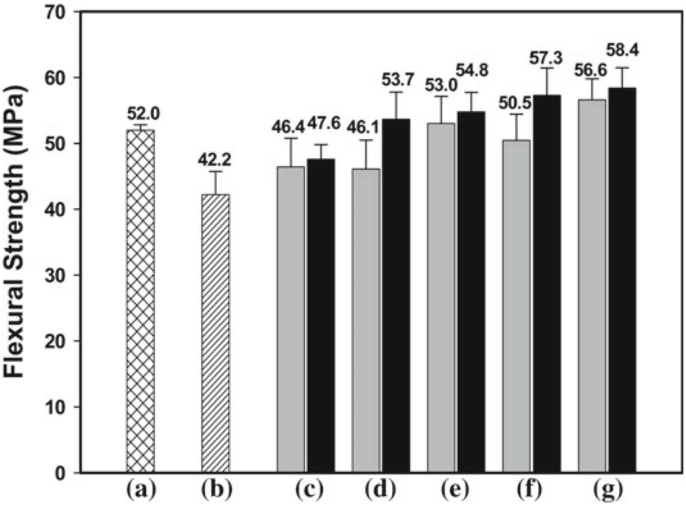


Fig. 9 Flexural strengths of **a** PP control, **b** untreated henequen/PP bio-composites, **c** treated by static (grey) and dynamic (black) methods with tap water, **d** NaOH 1 wt%, 10 min, **e** 1 wt%, 60 min, **f** 6 wt%, 10 min, and **g** 6 wt%, 60 min

of 1.1 g was 87 mg. It was concluded that the different flax-fiber separation processes lead to the verities size of fibers, which also display different surface characteristics.

Xue and Miao (2012) show a positive result on flax fiber treated with acetic anhydride, vinyltrimethoxy silane and NaOH. With 8 wt% concentration, the flax non-woven mat was first dipped in NaOH solution for 2 h at room temperature at 25 °C. The sample was then properly washed in cold tap water and neutralized with 5 wt% acetic acid solution for 30 min. It was then washed off in boiling water for 30 min and washed again thoroughly in cold tap water. The flax non-woven mat was dipped into a beaker containing acetic anhydride for 1 min and were drained for 3 min. The sample was then placed in a preheated oven (120 °C) for 120 min, which the time reaction was assumed to be completed. After the reaction, the samples were dried in vacuum oven at 120 °C. Vinyltrimethoxy silane was dissolved in ethanol/water (60/40 volume ratio) solution. The pH of the solution was adjusted between 3.5 and 4.0 by adding acetic acid. The flax non-woven mat was immersed in the solution (2.5 wt% silane with respect to the mat weight) at 25 °C for 1 h, followed by washing and rinsing it in distilled water. The treated sample was dried first in air at room temperature and then in oven at 105 °C for 6 h. Figure 10 shows the effect of different treatments permeability of the fibers.

Due to the changes in the surface polarity of the flax fiber, moisture absorption considerably reduced the resin flow permeability. Fiber–resin wettability was increased due to the alkalization treatment, which causes splitting of flax bundle fibers due to degumming and change in fiber orientation due to shrinkage of the non-woven mat. This leads to significant changes in-plane permeability and flow anisotropy of the treated non-woven mat. Relatively small scaled impacts were observed by the silane and acetylation treatments on in-plane permeability and flow anisotropy of the treated non-woven mat.

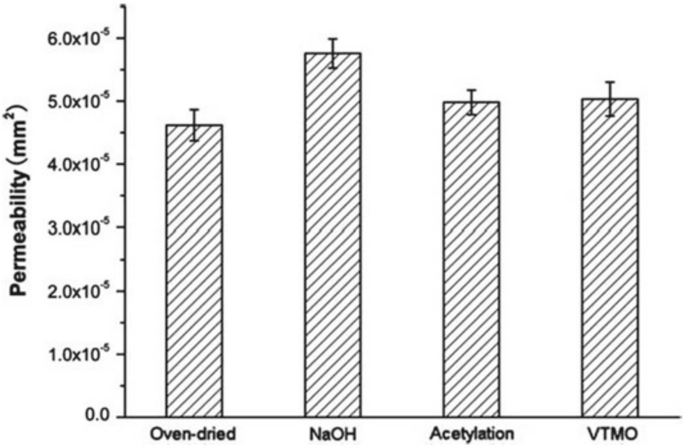


Fig. 10 Effect of different treatments on permeability

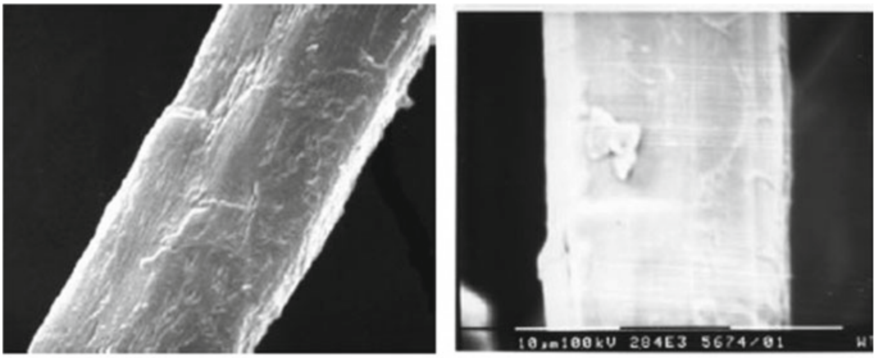
Wang et al. (2003) treated flax fiber with NaOH, benzoyl chloride, ethanol, dicumyl peroxide, acetone and alcohol. The fibers were first washed thoroughly with 2% detergent water and dried in an air oven at 70 °C for 24 h. The dried fibers were categorized as untreated fibers. Then another part of the flax fibers were subjected to sequential extraction with 1:2 mixture of ethanol and benzene for 72 h at 50 °C, followed by washing with double distilled water and air drying to remove waxes and water-soluble polymers before the chemical treatments. NaOH was used to pretreat the fibers.

Fibers were pre-treated with 5% NaOH for about half an hour to activate the OH groups of the cellulose and lignin in the fiber. The pre-treated fiber was dipped in alcohol water mixture (60:40) containing tri ethoxy vinyl silane coupling agent. The pH solution was maintained between 3.5 and 4, using the METREPAK Hydrion buffers and pH indicator strips. Fibers were washed in double distilled water and dried in the oven. An amount of pre-treated fibers were soaked in 18% NaOH solution for half an hour, filtered and washed with water. The treated fibers were suspended in 10% NaOH solution and agitated with benzoyl chloride. The mixture was kept for 15 min, filtered, washed thoroughly with water and filtered with filter papers before being dried again. For 1 h, the isolated fibers were then soaked in ethanol to remove the untreated benzoyl chloride and finally were washed with water and dried. After alkaline pre-treatments, fibers were coated with dicumyl peroxide from acetone solution. Saturated solution of the peroxide in acetone was used. To decompose peroxide, the favorable of all methods is set at high temperature. Figure 11 shows the flax fiber SEM.

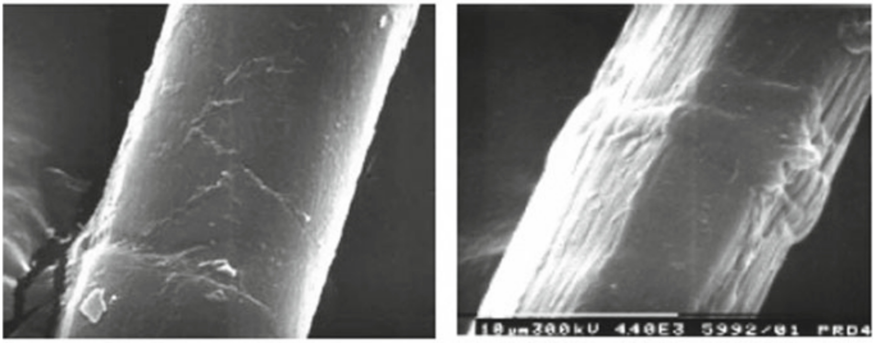
Wang et al. (2003) concluded that the surface properties and features were not clearly visible after the silane treatment. Due to the exhibited micropores on the surface of the flax fibers, a mechanically interlocking coating over the surface was a result of the coupling agent penetrating the pores. A major set of changes was observed on the fiber surface because of the benzoylation treatment. The presence of mass like substances accumulated over the surface of the fiber, smooth fiber surface was also observed. Meanwhile, dicumyl peroxide treatment shows that the surface topography was entirely modified and enhanced. The roughness of the fibers was witnessed by the micropores, particles that were seen adhering to the surface, groove like structures and protruding surfaces. Figure 12 shows the results of fiber bundle tensile strength test. It was clear from the result that silane treatment resulted in highest tensile strength of fiber compared to other treatments.

5.9 Agave americana *Leaves Fibers*

Thamae and Baillie (2007) treated *Agave americana* leaves fibers with NaOH and triethoxyvinylsilane. The leaves were cut from the central of the stocks. They were then boiled in water for 2 h, till the point it started to feel soft. The fibers were separated by gently squashing the leaves with rocks and the soft tissue was removed



(a) Untreated flax fibers (b) Silane treatment



(c) Benzoylation (d) Dicumyl peroxide treatment

Fig. 11 SEM results for chemically treated flax fiber

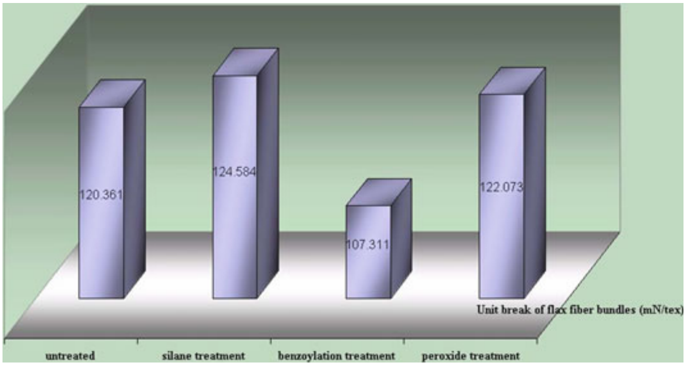


Fig. 12 Results of fiber bundle tensile strength test

by washing them with the water. The fibers were then subjected to drying, both air and sun, by hanging them in the open spaces along the fence.

Agave americana fibers boiled leaves were dipped into a 1M NaOH solution for 24 h. They were then washed in distilled water and dried in an oven for 24 h at 70 °C. Some of the mercerized fibers were dipped into a solution of 0.05% (w/w) triethoxyvinylsilane and agitated for 5 min. The solution was kept at pH 5.5 by acetic acid. Fibers were then washed and tried at 70 °C for 24 h. Fiber pull out test and tensile strength tests were carried out to examine the results which are shown in Figs. 13 and 14.

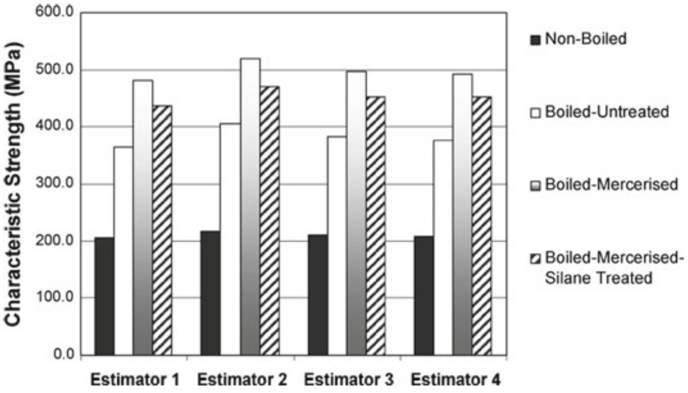


Fig. 13 Different tensile strengths for differently treated *Agave americana* leaves fibers

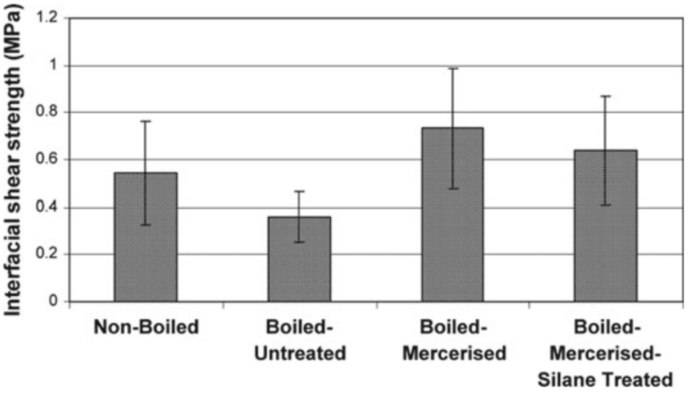


Fig. 14 Difference in interfacial shear strength of differently treated fibers

5.10 Kapok and Jute Fiber

Mwaikambo and Ansell (2002) treated kapok and jute fiber with NaOH and acetic anhydride. The fibers were first soaked in cold distilled water for 48 h at 20 ± 2 °C to eliminate any surface impurities. Fibers were then soaked in beakers containing different NaOH concentrations and were kept in a water bath maintained at 20 °C for 48 h. The fibers were then removed, washed with distilled water containing 1% acetic acid to neutralize excess sodium hydroxide and were then carefully rinsed with distilled water. Fibers were then treated with acetic anhydride containing concentrated H_2SO_4 as a catalyst for 5 min. Fibers were then washed with distilled water and dried. SEM was employed to evaluate the surface characteristics of the treated kapok and jute fiber. It was analyzed that the kapok fiber were not affected with chemical treatment, while jute fiber looked cleaner and refined along with the fiber surface. Figure 15 shows the untreated and treated jute fiber, while Fig. 16 shows the untreated and treated kapok fiber.

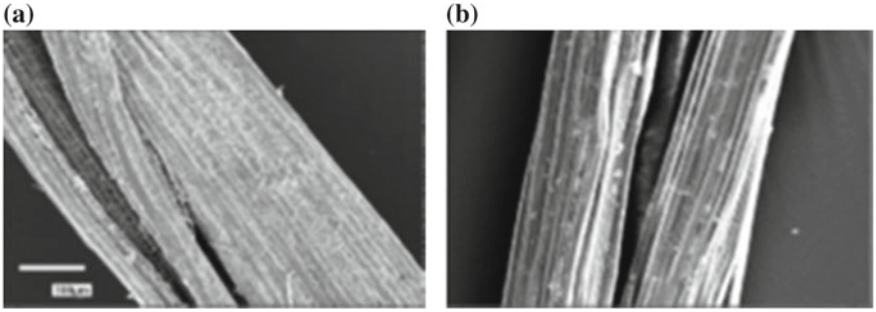


Fig. 15 **a** Untreated jute fiber, and **b** treated jute fiber

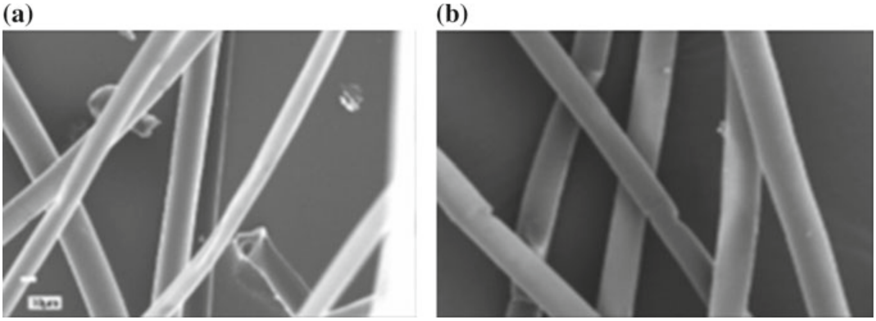


Fig. 16 **a** Untreated kapok fiber, and **b** treated kapok fiber

5.11 Acacia Wood

For unmodified Acacia wood fibers, it was washed and rinsed with distilled water. Later, it was dried in an open ventilated oven at 80 °C for 24 h. This process involved the use of two different man-made bio-based chemicals. The Acacia wood fibers were soaked for 24 h inside 1.5 L of 5 wt% of acetic acid solution (CH_3COOH) mixed with distilled water. After that, 90 g of sodium bicarbonate ($NaHCO_3$) was poured into the soaked wood for 24 h. The short Acacia wood fibers were filtered and rinsed using distilled water to neutralized remaining chemically attached to the wood fibers. Lastly, it was dried in an open ventilated oven at 80 °C for 24 h.

It is noticed that, Acacia woods have unique small porous structure, pits, opening, membranes, and an intricately layered wall. It tends to allow polymer matrices to enter in, which created a unique interlock between two or more molecules or particles. Alkaline treatment causes the wood fibers to be preserved and increase in pore size, structure, opening, pits, and membranes, which is due to the removal intricate layer wall of hemicellulose, lignin, wax, cellulose, and oil structure (Bakri et al. 2018). Thus, this increases the absorption rate of polymer into the wood structure, which was due to hydrophilic and hydrophobic interaction.

The bubble structure may be due to overheat, while the crack usually due to indirectly permissibility force during removal of composite sample. The addition of acacia wood sawdust chip particles creates an interlocking structure between two polymer blends in the composites. However, the untreated one have disadvantages than the treated one which cause poor surface adhesion. The treatment also creates surface roughness on the acacia wood sawdust chip particles. Gerard and Budtova (2011) shows similar results in their optical micrograph, where PLA/PHBV shows continuous structure that bind together.

6 Summary

It was learnt that chemical treatment can altered the mechanical properties of wood, natural fiber and hence the composite itself. It was also learnt that not all the natural fibers are affected by chemical treatments, some show inert behavior when treated with chemicals like kapok fiber. The study shows that not all the mechanical properties improve after the treatment of fiber, some of the mechanical properties have been affected in opposite way while some of the properties improved. Overall, the change in properties after the treatment of natural fibers might achieve a better performance as compared to untreated one. Many of the chemicals were employed by different researchers like Na_2CO_3, NaOH and K_2CO_3 because NaOH and K_2CO_3 showed great tendency to unveil the lignin structure in wheat straw, which makes good adhesion with polymer.

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Optimization of Fabrication Technique to Prepare Acacia Wood Reinforced Bio-composites

Md Rezaur Rahman, S. Hamdan, S. N. L. Taib and R. Baini

Abstract This chapter discuss the preparation and optimization of wood polymer composites based on the impregnation by polymer and nanoclay. Wood impregnation is one of the basic and most frequently used techniques to enhance the wood properties. This fabrication technique offers a wide range of applications depending on type of impregnants applied. Impregnation could make the wood less flammable, more dimensionally stable, more resistant to decay, harder, stronger, and more stable against UV rays. Softwood (Acacia) was impregnated with acrylonitrile, poly(vinyl) alcohol and organically nanoclay. The specimen preparation was carried out using the vacuum-chamber in a laboratory scale. The physical and mechanical properties of the modified wood were analyzed through Tensile and Flexural tests, SEM, FTIR, TGA and DSC. Mechanical test results shown that Tensile and Flexural strength have improvements with the addition of the nanofillers. The FTIR test shown that the chemical bonding between PVA into the wood cell would certainly enhance the matrix adhesion and contribute to its property enhancement. SEM illustrate the samples surface morphology which confirm the impregnation of the specimen. TGA results shown the additives impregnate into the wood component increase the thermal stability compared to the raw wood. DSC results indicate the impregnate wood has a higher melting temperature compared to the raw wood, due to existing of the polymer and nanoclay interfacial bonding among cell wall of the wood. Response surface methodology (RSM) was used to optimize the conditions for the preparation of wood composites. The design experiment was carried out using Design Expert 11.

Keywords Wood · Bio-composites · Modification · Reinforcement

1 Introduction

To date, crude oil-derived composites have been commercially used in varies application (Lizasoain et al. 2015). However, arising environmental awareness and new

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Md R. Rahman (✉) · S. Hamdan · S. N. L. Taib · R. Baini
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa,
94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

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rules and regulations are forcing industries to seek more ecological friendly materials for their products (Rahman et al. 2010). New generations of bio-based building materials made from renewable resources, such as wood are becoming more interesting. Products that made up of thermoplastic such as polyethylene and polypropylene are traditionally used long time ago. Wood is made of cellulose, hemicellulose and lignin that are multifaceted structures which are biologically originated (Rahman et al. 2013). Wood is a natural product which is susceptible to biodegradation, weather conditions, chemical degradation, fire and mechanical wear. These processes lead to the change of the wood properties such as weight, dimensions and color. There a lot of techniques to protect wood against these biotic and abiotic factors. Treatments of the wood cell wall offers a wide range of protection depending on used impregnate substances. In addition, impregnation can make the wood more resistant to decay, less flammable, more dimensionally stable, harder, stronger, more UV stable resistant and high resistance to moisture or water.

Wood has a diversity of application such as construction, furniture, home decorating, and tools due to its gorgeous features. Solid wood has such high-physical strength, low processing cost and attractive desirability that make them so attractive for building and construction application (Rahman et al. 2013). However, the source of the hardwoods was decreasing due to the slow of growing rate of the hardwood plant. So, researchers and manufacturers find an alternative way to replace the source of hardwood by modify the properties of the softwood and low-density wood to have the high-quality of hardwood. To achieve the objective, appropriate test to increase the extent of wood properties such as physical properties, thermal stability, durability, decay resistance, water resistance and hardness to meet necessity of end-use application.

A new material has been spawned, which is combination of a thermoplastic component and wood-based component know as wood plastic composites (WPCs). Over the years, the wood has been reinforced with many types of chemicals such as styrene, epoxy resins, phenol, formaldehyde and vinyl to change its physical and mechanical properties. According to Fabiyi et al. (2009), WPCs are combination of wood and chemical additives such as plasticizers, lubricants, coupling agents, nucleating agents, pigments and UV stabilizers. Consequently, the use of additive agents can improve certain properties of wood material especially the flexural, tensile, durability, relative to pure wood (Binhussain and El-Tonsy 2013). Also, modified wood is more resistive against water absorption and high resistance to decay, insects and ultraviolet damage (Rahman et al. 2010). However, there are disadvantages of the WPC which is the minor change of the natural color on the modified wood.

The concept to produce WPC is straightforward. The sawdust, wood shavings or woody biomass from agricultural residues is blended with polymers such as polyethylene or polypropylene within an extruder or through injection molding (Kristoffer 2012). The replacement of the WPC over the plastic material in application is contribute to the development of green chemistry by increasing usage of biodegradable material and reuse of waste generated from wood industries (Martins et al. 2017). Moreover, usage of WPC promotes the awareness of environmental issues. The report by Martins et al. (2017) stated that, the footprint of CO_2 of WPC that submitted to combustion or landfill disposal is neutral.

The great evolution of the WPCs has risen together with new development of processing technologies which lead the demand on WPCs to be increased (Martins et al. 2017). The popularity of WPC is also due to high durability, low maintenance and reasonable price to other competing materials. In addition, WPCs becomes fully natural material if the polymer used is biopolymer based such as polylactic acid (PLA) and polyhydroxyalkanoates (PHA) (Martins et al. 2017). The usage of WPCs is varied in application during the past decade and become widespread in many building applications such as decking, docks, fencing, landscaping and home interior and exterior (Yadav and Yusoh 2016).

2 Literature Review

2.1 Polymer Composites

Composites contain two materials or phases which aim to propose a combination of mechanical and physical properties and applications (Mohanty et al. 2002). As an example, matrix and filler called a reinforcement or predominantly dispersed phase and sometimes additional compound are being added (Barton et al. 2014). Matrix will allow shaping products appropriately (Barton et al. 2014). The grossing demand of polymer matrix composites (PMCs) is due to the several factors such as the low cost of consumption and better properties (Mohanty et al. 2002). The matrix in PMCs could be produced from duroplastic or thermoplastic polymers (Mohanty et al. 2002). Several biological materials may be employed into the polymer materials. According to Roy (2014), they believe that polymer bio-composite materials will reduce the need for synthetic production, thereby producing a positive effect for environmentally and economically. In the extensive research, the properties of polymer composite produced with plant materials depend mostly on inter-phase interaction which is hydrophobic matrix and hydrophilic filler (Barton et al. 2014).

2.2 Bio-composites

Composites made up of two or more distinct constituents which when together combined will give entirely different properties form the individual components (Roy 2014). Polymer composite that contains one component of bio-based and biodegradable are called bio-composites (Barton et al. 2014). Eco-friendly composite is a similar name to describe the bio-composites. The bio-composites contain large of organic and inorganic components, such as natural and synthetic polymers, polysaccharides, proteins, sugars, ceramics, metals and nanocarbons. Many studied have

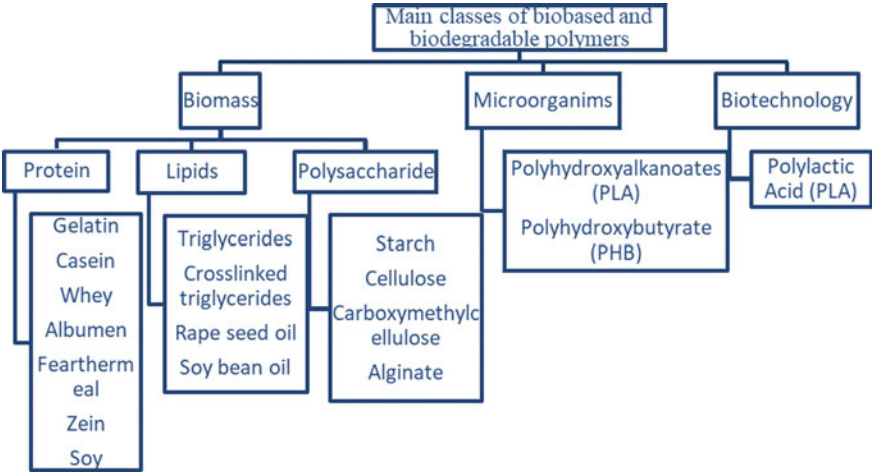


Fig. 1 Different classes of polymers which are bio-based and biodegradable (Bugnicourt et al. 2014)

been carried out to investigate the mechanical properties and physical properties to improve the functionalities of material to achieve better result. The bio-composites shows the growing interest and been developing for quite some time, due in increasing price of oil and oiled-derived products together with the after service environmental pollution (Faruk et al. 2014). They also give a nice natural look, grip to composite and not harmful to regarding environmental impact (Mohanty et al. 2002). The above characteristics of bio-composite completely explained eco-friendly products show more attraction is daily use.

Furthermore, natural fiber is the main source in bio-plastic production because of the good mechanical properties tested to compete with glass fiber product (Faruk et al. 2014). Moreover, due to characteristics that it has, bio-composites attracted for some application such as bio-degradable packaging (Faruk et al. 2014). Additionally, bio-composite shows good result in bio-degradability in the sake of ease recycling (Faruk et al. 2014). Figure 1 shows the classification of bio-based and biodegradable polymers.

2.2.1 Bio-matrix Based Polymer Composites

The term bio-matrix described the composite matrix is partly or wholly biodegradable (Mohanty et al. 2002). A solution to waste disposal problems of plastic via petroleum is by the turn out the market scenario to use of biopolymers (Lizasoain et al. 2015). Moreover, the biodegradable polymer is defined polymer that capable of undergoing decomposition, primarily through the enzymatic action of microorganisms into CO_2, methane inorganic compounds or biomass within the time required (Mohanty et al. 2002). Another definition state, the polymer that can adapt to undergo microbially induced chain scission, leading to mineralization, oxidation, photodegradation and which can change the polymer during decomposition process (Mohanty et al. 2002). These polymers are categorized by the scientist into three types which are bio-synthetic, semi bio-synthetic and chemosynthetic biopolymers (Lizasoain et al. 2015). Bio-synthetic can achieve full degradation according to the time scale due made up of renewable source (Lizasoain et al. 2015). Also, for semi-biosynthesis and chemosynthesis are showing good performance in bio-degradability because both contain naturally occurring chemical bonds (Lizasoain et al. 2015). From the categories describe can all of them capable to function as thermosets and thermoplastics (Lizasoain et al. 2015).

2.2.2 Natural Fiber Reinforced Composites

The uncertainty behavior of the petroleum oil prices and their resource led the consumer to find another alternative to achieve the market demand which is natural fiber reinforced polymer (Faruk et al. 2014). Natural fibers are divided into three groups which group of animals, mineral and plant or vegetable (Lizasoain et al. 2015). Variety group of natural fibers increase adhesion to the matrix and makes them an attractive gap-filler material (Roy 2014). The different types of natural fiber show the difference of their capability so vary tested were employed to checking the natural fiber reinforced composites performance (Faruk et al. 2014). As a fiber, it will change the mechanical and physical properties when reinforcement it takes part in the composite materials (Roy 2014). Moreover, when fiber-reinforced composite new strength and stiffness are introduced, so new suitable structural composites can be made (Roy 2014). Apart from improving adhesion, reinforcement also aims to limit the water adsorption, increasing dimensional stability and increasing resistance against environmental factors such as temperature (Barton et al. 2014).

2.2.3 Application of Bio-composites

Bio-composites are created by incorporating natural fibers into green polymer materials. Uses of bio-composites promoted the environmental advantages such as reduction of fossil fuel use and lower the green-house gas emissions (Roy 2014). Examples of bio-composites include starch, cellulosic, soy-based plastic, polylactides, soy-based resins (Roy 2014). The main markets that gross the demand of bio-composites is construction and automotive sectors (Roy 2014). Further improvement is carried out to increase performance of bio-composite to ensure it is working perfectly on every application (Mohanty et al. 2002). So, reinforcing the fibers such as cotton, jute, flax, hemp, sisal and corn are increasing as substitution of synthetic fibers. Natural fiber reinforced polymeric composites shows an excellent result when employed to automobile parts such as doors and body panel of the cars (Uddin and Kalyankar 2011).

However, before the present of bio-composite, a lot of non-recyclable and non-biodegradable product is made up for automotive industry and it give major environmental impacts such as greenhouse emission (Roy 2014). Then, because of the bad impact towards the environment, automotive industry has increased the use of natural fiber composites on their several components, such as floor well panel and foot well panel (Faruk et al. 2014). According to Faruk et al. (2014), several automotive components are made up of natural composites which generally based on polypropylene resin and fibers such as flax, hemp, kenaf and sisal. Moreover, the components such as door panels, package trays, seat backs are produced using natural fibers reinforce thermoplastics and thermosets (Faruk et al. 2014). According to Roy (2014), the components made up of natural fiber composites are much lighter than metal base car components and contain the same strength and more reliable because is non-corrosive (Roy 2014).

In addition, bio-composites are widely used in the construction sector (Faruk et al. 2014). In the construction industries, they have the potential to replace the wood and oriented strands boards (OSB) laminates in the structural insulated panels (SIPs) (Uddin and Kalyankar 2011). The manufacturing and structural feasibility of natural fiber reinforced polymeric structural insulated panels for panelized construction (Roy 2014). They showed a good performance over traditional wooden and concrete construction (Uddin and Kalyankar 2011). Moreover, composite reinforced these fibers require low cost in the application, require less energy for manufacturing and do not affect adversely the manufacturing tools (Uddin and Kalyankar 2011). Furthermore, one of the reasons natural resources composites are chosen is because of the low price, weight reduction and marketing incentives (Roy 2014). These features have attracted much attention to various sector to change to the bio-composites. Also, due to government support in term of environmental advantages and availability of the resources (Roy 2014).

2.2.4 Potential Market Demand Toward Bio-composites

From the study by Faruk et al. (2014) on 2010, they displayed the total global natural fiber composites materials market shipments topped to 430.7 million pounds with a value of US$ 289.3 million and market is expected to grow to US$ 531.3 million in 2016 with an 11% Compound Annual Growth Rate (CAGR) over the next 5 years. The development in emerging bio-based plastic showed the positive result in the market place with an average annual growth globally was 38% from 2003 to 2007 (Faruk et al. 2014). In the Europe market, the percentage of annual growth seems higher than global result in the same period which is 48% (Faruk et al. 2014). The worldwide capacity of bio-based plastics is expected to produce 3.45 million metric tons in 2020 from 2.33 million metric tons in 2013 (Faruk et al. 2014). Due to that situation, economists predicted that the worldwide capacity of bio-based plastics is expected to produce 3.45 million metric tons in 2020 from 2.33 million metric tons in 2013 (Faruk et al. 2014). While in the automotive sector total consumption of plant materials in 2012 in Europe amount to 80-kilotons (Barton et al. 2014). From the review consumption of product based on plant, materials receive massive attention globally.

2.3 Acacia Wood

Acacia mangium and *Acacia auriculiformis* are sub-family species of *Mimosoidee*, which are the leguminous tree. Mainly, native to north Queensland, Australia, the trees are also found in Papua New Guinea and Moluccas Islands of Indonesia (Bakri et al. 2018a, b). These species have been chosen as plantation species due to their fast growth, good form and utilization potential of the timber. About one million hectares of lands in Sarawak are reserved for plantation and reforestation of acacia trees and other species for timber industry purpose (Bakri et al. 2018a, b). In addition, the other aim of reforestation is to conserve natural forest as well and animal habitats (Bakri et al. 2018a, b). In Sabah, acacia trees function as a firebreak in 1967 (Bakri et al. 2018a, b). Moreover, in Bangladesh, *Acacia auriculiformis* have been sort listed as the priority species from the Bangladesh Forest Department. Therefore, this species is very multipurpose trees it is functioned to provide shades from windbreaks and reduce soil erosion in agroforestry systems (Professors 2009). Furthermore, three countries of the Southeast Asia which are Malaysia, Indonesia and Thailand have a massive plantation of acacia trees due to the demand of wood, pulps, paper mills and furniture industries (Rokeya et al. 2010).

Acacia mangium species is susceptible to the heart rot damage due attacked of fungal and bacteria to the wood which causes the decay at the center of the wood (Bakri et al. 2018a, b). So, the stiffness of the wood will increase (Bakri et al. 2018a, b). To overcome this complication, new *Acacia mangium* hybrid plant are created. The hybrid acacia is the cross of two species which are *Acacia mangium* and *Acacia auriculiformis* (Bakri et al. 2018a, b). Hybrid acacia bole is long and straight with light branches (Bakri et al. 2018a, b). The production and growth rate of hybrid acacia is more rapid than the other species and it is more resistance towards heart rot disease (Rokeya et al. 2010). The physical properties of hybrid acacia in different seasoning condition shows different result (Rokeya et al. 2010). From the test carried out, the toughness tangential of hybrid acacia in the green condition is higher than in the air-dry condition which 187 and 90 respectively (Rokeya et al. 2010).

2.3.1 Physical Properties of Acacia Wood

The major physical properties of Acacia Wood are measured based on the radial, tangential, volumetric shrinkage and density (Bakri et al. 2018a, b). *Acacia auriculiformis* has the highest of basic density, followed by acacia hybrid, *Acacia mangium* and 2nd Generation *Acacia mangium*. Due to lower density, 2nd Generation *Acacia mangium* shower the fastest growth than *Acacia mangium* itself. In terms of volumetric shrinkage, *Acacia auriculiformis* showed the highest than the other species (Bakri et al. 2018a, b). Percentage of wood shrinks which contain high density was about 3.30%. According to Rokeya et al. (2010), the volumetric shrinkage and specific gravity of acacia hybrid in green and air-dry condition were around 9.71 and 0.56% respectively. The acacia wood may be shrinking more when it contains high density (Bakri et al. 2018a, b). In addition, the specific gravity of the Acacia Hybrid in age four years in Malaysia showed around 0.51 (Bakri et al. 2018a, b). The specific gravity of the acacia wood may depend on the fiber wall thickness in diffuse-porous hardwoods (Bakri et al. 2018a, b). According to Bakri et al. (2018a, b), the specific gravity for *Acacia auriculiformis* and *Acacia mangium* are 0.61 and 0.52 respectively.

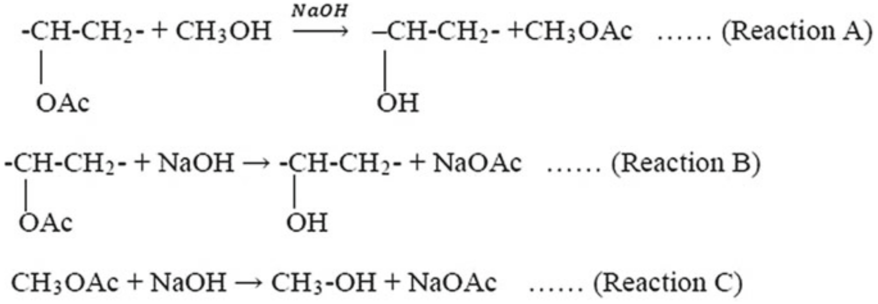
2.3.2 Mechanical Properties of Acacia Wood

To identify the mechanical properties of acacia wood, some test need be run to understand the stiffness, tensile strength, crushing strength, resilience and shearing strength (Bakri et al. 2018a, b). According to Bakri et al. (2018a, b), the Modulus of Rupture (MOR) and Modulus of Elasticity (MOE) of acacia hybrid with four years age was 713.47 and 82.83 kg/cm^2. In addition, Bakri et al. (2018a, b) documented the modulus of rupture and modulus of elasticity of acacia hybrid was found greater than *Acacia auriculiformis* and *Acacia mangium* which are 658 and 652 kg/cm^2; 83 and 79 kg/cm^2, respectively. While, the 2nd Generation of *Acacia mangium* has a slightly lower value of MOR and MOE than the *Acacia mangium*. Also, they showed the different result of strength value that performed on four years old acacia hybrid and six years old *Acacia mangium*. From the result, relation shows that low strength was affected by the low density (Bakri et al. 2018a, b). The report showed the MOE and MOR value of *Acacia auriculiformis* was 652 and 79 kg/cm^2, lower than *Acacia mangium*, which found 658 and 83 kg/cm^2 respectively (Bakri et al. 2018a, b). From the research on 4-year-old acacia hybrid, the value of MOR and MOE is 82.83 and 713.47 kg/cm^2 respectively. However, from the detailed of various researcher strength value of acacia hybrid are not consistent. It may different due to genetic, age, growth rates and geographical factors. But in generally, all the tested species are found to have satisfactory performance for different purpose and application (Bakri et al. 2018a, b).

2.4 Polyvinyl Alcohol

Polyesters are the groups of polymers in which each monomer are bonded via ester linkages. Polyvinyl alcohol is known as water-soluble polyhydroxy polymer, based on its easy preparation, nontoxic, noncarcinogenic, biodegradable and bio-adhesive characteristics, excellent chemical resistance and physical properties (Zhu et al. 2013). In some research, PVA shows an excellent as biomaterials due capable of simulating natural tissues, contact lenses, the lining for artificial organs and drug delivery (Zhu et al. 2013). However, PVA had some limitation in high hydrophilicity but it can hold good mechanical properties on the dry state (Zhu et al. 2013). Back to the history, Herrmann and Haenhnel are the first to PVA by saponifying poly(vinyl esters) with caustic soda solution in 1924 (Hallensleben et al. 2015). They used the saponification process of poly(vinyl acetate) (PVAc) to synthesize PVA. Thus, in 1924 both discovered new techniques to prepared PVA by transesterification with absolute alcohols in the presence of catalytic amounts of alkali in 1932 (Hallensleben et al. 2015). Preparing the PVA cannot directly undergo polymerization to tautomerism of vinyl alcohol monomer (Lyoo and Lee 2002). This is due to vinyl alcohol which is the monomer is unstable with respect to acetaldehyde (Hallensleben et al. 2015). Some of the methods can be accomplished, where PVA is synthesized by the saponification of a poly(vinyl ester), such as poly(vinyl pivalate) and (PVAc) (Lyoo and Lee 2002). Other precursor polymers used are formatted group and chloroacetate groups instead of acetate (Hallensleben et al. 2015). The saponification mechanism was interpreted according to the following chemical reaction as shown in Fig. 2.

Alcoholysis of polyvinyl acetate is shown in the Reaction A. Then, follow by Reaction B where saponification of PVAc and production of methyl acetate in reaction C. PVA consists mainly of 1,3-diol units and PVA contains 1,2-diol units that obtained by hydrolysis of PVAc usually in range of 1–2% (Hallensleben et al. 2015). A high degree of 1,2-diol units poses change of some properties for example degree of swelling of films in water (Abdullah et al. 2017). However, a high degree of 1,2-diol can be reduced by lowering the temperature during polymerization process of vinyl acetate or using another vinyl ester such as vinyl format and vinyl benzoate (Hallensleben et al. 2015). In industries, acetylene and ethylene are used as base materials to produce vinyl acetate in the presence of acetic acid and oxygen. Thus, purified vinyl acetate induce heat to initiate the polymerization process with methanol solution (Abdullah et al. 2017). According to Abdullah et al. (2017), PVAc is produced from about 70% of monomers during the polymerization to use for preparing PVA via saponification with caustic soda. Figure 3 shows the flowchart of PVA industrial manufacturing (Abdullah et al. 2017).



$$-\underset{\mid \atop \text{OAc}}{\text{CH}}-\text{CH}_2- + \text{CH}_3\text{OH} \xrightarrow{NaOH} -\underset{\mid \atop \text{OH}}{\text{CH}}-\text{CH}_2- + \text{CH}_3\text{OAc} \quad \ldots\ldots \text{ (Reaction A)}$$

$$-\underset{\mid \atop \text{OAc}}{\text{CH}}-\text{CH}_2- + \text{NaOH} \rightarrow -\underset{\mid \atop \text{OH}}{\text{CH}}-\text{CH}_2- + \text{NaOAc} \quad \ldots\ldots \text{ (Reaction B)}$$

$$\text{CH}_3\text{OAc} + \text{NaOH} \rightarrow \text{CH}_3\text{-OH} + \text{NaOAc} \quad \ldots\ldots \text{ (Reaction C)}$$

Fig. 2 The saponification mechanism according each chemical reaction (Abdullah et al. 2017)

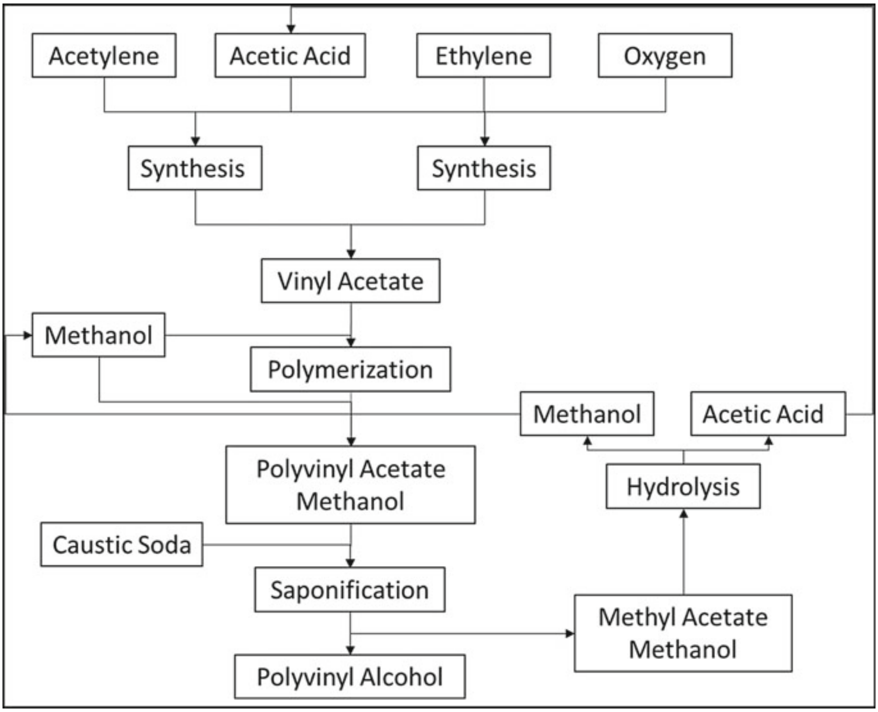


Fig. 3 Flow chart of PVA industrial manufacturing (Abdullah et al. 2017)

Physically, PVA is large crystalline lattice modulus, due to these characteristics PVA fibers and film have high tensile modulus and good abrasion resistance (Lyoo and Lee 2002). Moreover, the physical properties can be maximized by increased the degree of saponification, the molecular weight and the diotacticity. The different tacticity of PVA will have different properties (Hallensleben et al. 2015). Improvement of four polymerization method of vinyl acetate affects the molecular weight eventually to physical properties. Four polymerization method are bulk, polymerization, solution polymerization, emulsion polymerization and suspension polymerization (Lyoo and Lee 2002; dos Reis et al. 2006). From the study of the researcher, suspension polymerization of vinyl acetate is possible to reach high conversion compare to other polymerization methods (Lyoo and Lee 2002). The acetyl groups of partially saponified PVA can be distributed statistically depending on the condition of production. The distribution is significant to influence the properties of PVA such as melting point, surface tension of aqueous solutions and the protective colloid properties (Hallensleben et al. 2015). Figure 4 shows the PVA molecular structure fully-hydrolyzed.

Commercially polymer and copolymer are accomplished by suspension polymerization. Lyoo and Lee (2002), state the highest conversion of molecular weight and polymerization rate can be done throughout the suspension polymerization compare

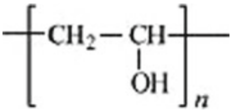


Fig. 4 PVA molecular structure fully-hydrolyzed (Abdullah et al. 2017)

to other polymerization techniques. The polymer that is produced by suspension polymerization generally with diameters in the range of 80–700 μm (Lyoo and Lee 2002). Moreover, this method of suspension polymerization can reduce the exothermic heat and prevent viscosity enhancement (Lyoo and Lee 2002). Besides, the properties of PVA depends on their molecular mass and residual content of acetyl groups which have mentioned before. In the industrial production process, the process to produce polyvinyl acetate for processing PVA is polymerization in methanol. So, the polymerization temperature, vinyl acetate-methanol ratio and polymerization conversion are the factor determining the molecular mass of PVA (Hallensleben et al. 2015). Reducing the polymerization temperature, the proportion of methanol and the conversion will increase the molecular mass. Besides that, the hydrolysis condition influences the forming of PVA. Also, the content of residual acetate group can be adjusted with varying the amount of catalyst, temperature and reaction time.

2.4.1 Properties of Polyvinyl Alcohol

PVA consists of the aliphatic backbone where makes it slow to degrade. However, the basic properties of PVA depending on the degree of polymerization and degree of hydrolysis (dos Reis et al. 2006). Most of PVA production divided into two different degrees of hydrolysis which are 98 mol% of acetyl groups and those with the degree of hydrolysis of 87–89 mol% (Hallensleben et al. 2015). Both of PVA products have a degree of polymerization in between of ca. 500 and 2500 (Hallensleben et al. 2015). The commercial PVA normally is white to yellowish and manufactured as powder and granules (Hallensleben et al. 2015). Manufacturers usually characterized the PVA based on the molecular weight of precursor polymer and the degree of hydrolysis. Hallensleben et al. (2015) states, the melting point of fully saponified PVA and glass transition temperature are 228 and 85 °C respectively. Both conditions are depended on the distribution of acetyl group, tacticity and water content on PVA.

Besides that, the viscosity of an aqueous solution of PVA was relying on the concentration, degree of polymerization and degree of hydrolysis as well (Hallensleben et al. 2015). PVA that fully saponified shows more viscous than the partially saponified PVA even at the same degree of polymerization. As mention before a tensile strength of PVA relies on molecular weight, degree of polymerization of precursor polymer and degree of hydrolysis. Form the study by Abdullah et al. (2017), full hydrolysis of PVA possessed a tensile strength of 1.6 GPa, Young's Modulus of 48 GPa and elongation break of 6.5%. While, for partial-hydrolysis PVA shows a slightly lower than the full hydrolysis PVA which are 25.4 MPa, 27.6 MPa and 260% for tensile strength, Young's Modulus and elongation break respectively (Abdullah

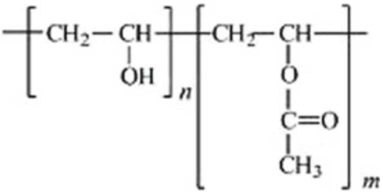


Fig. 5 PVA molecular structure partially-hydrolyzed (Abdullah et al. 2017)

et al. 2017). Furthermore, the mechanical properties of PVA can be improved when crosslinked with related chemicals such as boric acid, citric acid, and hexamethylene diisocyanate (Abdullah et al. 2017). Also, the addition of polyol that acts as plasticizers could improve the elongation break due to the increase in mobility of polymer molecules (Abdullah et al. 2017). According to Abdullah et al. (2017), mechanical properties PVA blend shows improvement with crosslinking with sodium benzoate as a crosslinking agent with the aid of ultraviolet irradiation. Figure 5 shows the PVA molecular structure partially-hydrolyzed.

Thermal properties of biodegradables polymers are very significant to study for use of the application. In the extensive research, according to the thermogravimetric analysis (TGA), the melting temperature and glass transition temperature for pure PVA is 230 and 85 °C respectively (Abdullah et al. 2017). Basically, the glass transition temperature is depending on the degree of hydrolysis of PVA. Abdullah et al. (2017) reported, the glass transition temperature for 98–99% hydrolyzed PVA and 87–89% hydrolyzed PVA is 85 and 58 °C respectively (Abdullah et al. 2017). From the data, the glass transition temperature of PVA can be summarized increasing linearly with decreasing relative humidity due to the plasticizing effect of water on the PVA crystalline structure (Abdullah et al. 2017).

PVA is known as the biodegradable polymer but some of the necessary condition is not completely biodegradable due to missing types of microorganisms, changes of temperature, pH level and humidity (Abdullah et al. 2017). PVA is highly biodegradable in activated sludge, instead biodegradation rate is very low in environment condition such as compost and soil (Abdullah et al. 2017). Biodegradation rate of PVA might be lower than the PVA blends with other biopolymer such as lignocellulose and gelatin due to lack of microorganisms to attack pure PVA. From the study of Abdullah et al. (2017), found the degradation of PBA based films of 8–9% required 74 days compared to 13% which only 21 days. Therefore, PVA blended with biopolymer such as starch and cellulose increase the biodegradation rate.

2.4.2 Application of Polyvinyl Alcohol

Many types of synthetic polymers have been used widely in variety of application such as pharmaceutical and biomedical application especially for controlled drug release tests due to its non-toxic and degradable properties (dos Reis et al. 2006). Also, PVA has been used in textile and paper industry applications (dos Reis et al. 2006). In the paper industry, PVA is used to coated papers to create specific barrier properties.

Also, proven act as pigment binding and excellent carrier for optical brighteners (Hallensleben et al. 2015). Moreover, PVA act as sizing agent to emphasize the surface has sufficient adhesion (Zhifeng 2007). So, with enough of adhesion fiber in warp yarns and non-woven fabric can attach perfectly and will have a good strength. Zhifeng (2007) reported, the correlation of PVA molecular structure and adhesive strength of PVA to polyester and cotton fibers are leys on the degree of polymerization and degree of hydrolysis. From their research, shows the maximum strength of PVA increase with the increase of the degree of polymerization and degree of hydrolysis (Zhifeng 2007). In partially alcoholized PVA, a certain amount of acetyl groups is present. Thus, it poses some changes to adhesives bonding such as decrease of adhesive bonding and decrease of internal stresses between the adhesive layer and fiber (Zhifeng 2007).

Furthermore, Polyvinyl alcohol (PVA) has widely been used as a crosslinking agent to produce biodegradable polymer matrix compounds (Islam et al. 2014). PVA has proven to be effective as protective colloid to produce plastic dispersions, especially as suspension stabilizer, PVA homo- and copolymers (Hallensleben et al. 2015). Moreover, PVA is very suitable for thermoplastic production of packaging films with graded water solubility (Hallensleben et al. 2015). The objective of reducing petroleum based plastic drive PVA as primarily based to produce degradable packaging in packaging applications (Abdullah et al. 2017). Moreover, the growing concerns of the environmental impact form non-degradable polymer give impact to replace with fully biodegradable plastic such as PVA, PLA and other types of aliphatic polyesters (Abdullah et al. 2017). Also, the main purpose of the packaging process to ensure the manufactured products in packages is save from the deterioration through transportation, storage and to prolong the life of the packaging as well.

2.5 Acrylonitrile

Acrylonitrile is known as an intermediate key in the chemical industry for the production of a wide range of products (Razdil et al. 2012). Also, name as 2-propenenitrile, acrylic acid nitrile, propenoic acid nitrile, vinyl cyanide and propylene nitrile. Acrylic fibers are revolved from polymers that are made from monomers containing 85% by mass acrylonitrile in the chain (Gupta and Afshari 2009). First preparation of acrylonitrile and polyacrylonitrile are found by a French Chemist, CH. MOUREAU, via the dehydrating process of acrylamide with phosphorus pentoxides in 1983 (Hansora 2014). However, the technique not commercialized for the industrial production and application. Acrylonitrile is accomplished commercially via the propylene ammoxidation process which known as SOHIO process (Razdil et al. 2012). The technique used for manufactured acrylic fibers is free radical technique (Gupta and Afshari 2009). Generally, the production of acrylonitrile is done by the reaction of propylene, ammonia and oxygen with the presence of a heterogeneous catalyst. Synthetic

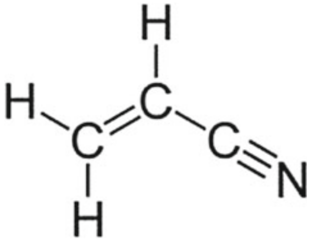


Fig. 6 Chemical formula of acrylonitrile (Razdil et al. 2012)

rubber-based acrylonitrile knew as acrylonitrile butadiene rubber (Gupta and Afshari 2009). Figure 6 shows the chemical formula of acrylonitrile.

The demand for acrylonitrile is increase when acrylic fiber called Orlon discovered by DuPont (Gupta and Afshari 2009). The second category of acrylonitrile-based fibers name as modacrylic fibers. This second type of modacrylic fiber contains about 35–85% by mass of acrylonitrile (Gupta and Afshari 2009). This category is very excellent in fire retardant because the comonomers used almost all based on halogenated ethylene which technically unsaturated molecules (Gupta and Afshari 2009). According to Gupta and Afshari (2009), modacrylic fibers must apply heat stabilization and stretching after spinning to develop necessary properties. As mention before acrylonitrile can be manufactured from several types of a chemical such as propylene, acetylene and ethylene. Furthermore, there are varies of polymerization technique in manufactured acrylonitrile such as solution polymerization, bulk polymerization, slurry polymerization and emulsion polymerization (Gupta and Afshari 2009).

2.5.1 Properties of Acrylonitrile

The individual polyacrylonitrile chain consists of highly polar nitrile groups and attaches to carbon atoms which make chain is strong enough. However, the existence of nitrile groups in adjacent arrangement could pose some repulsion on the chain (Gupta and Afshari 2009). This repulsion occurs due to the bond angles of the nitrile groups cannot accept the antiparallel arrangement (Gupta and Afshari 2009). Thus, makes the chain arrangement changes to an irregular helical form (Gupta and Afshari 2009). Acrylic and modacrylic fibers usually prepared as tow and staple for commercializing purposed with a small quantity of continuous filament sold. The linear density for the product produced is between 0.84 and 17 dtex (Gupta and Afshari 2009). According to the review by Gupta and Afshari (2009), the tensile strength of acrylic and modacrylic fibers are lower than the polyester and polyamide fiber but nearly to cotton and higher than wool. Razdil et al. (2012) stated, acrylonitrile is a clear colorless liquid at ambient condition and known as polar molecules due to the presence of the electronegative nitrile functional group that is conjugated with carbon-carbon double bond. The result of the fibers dipolar interaction between nitrile groups differentiate the physical characteristics of acrylic fibers such as high electrical resistance, moderate flammability, resistance to sunlight and resistance towards microbiological attack (Gupta and Afshari 2009). Besides that, due to polarity, it will poses moisture about 2–3% and will give the fiber relatively low value of glass transition temperature in a range of 70 °C (Gupta and Afshari 2009).

Acrylonitrile is very soluble in a variety of organic solvents such as acetone, benzene, carbon tetrachloride, diethyl ether, ethyl acetate, ethylene cyanohydrin, petroleum ether, toluene and methanol. Table 1 shows the common azeotropes of acrylonitrile (Razdil et al. 2012). Also, acrylonitrile is miscible in the water at a certain temperature. According to Razdil et al. (2012), the vapor pressure of acrylonitrile in aqueous solution increases gradually with the concentration. Moreover, the chemical reactivity can occur at two reactive sites which are the carbon-carbon double bonds and nitrile functional groups. Two principle reactions of acrylonitrile which are polymerization and dehydration (Razdil et al. 2012). However, there are others principle reaction like the addition of halogens across the double bond to produce dihalopropionitriles and cyanoethylation of alcohols. All these principles are used to produce end uses of acrylonitrile (Table 2).

In the study by Zhu et al. (2013) shows the tensile strength improvement of polyacrylonitrile (PAN) is proven with blend of PVA. The value of tensile strength of the polymer blend film is increased with increase of PVA mole contents in the sample.

Table 1 Azeotropes of acrylonitrile (Razdil et al. 2012)

Azeotrope	Boiling point (°C)	Acrylonitrile concentration (wt%)
Tetrachlorosilane	51.2	11
Water	71.0	88
Isopropyl alcohol	71.7	56
Benzene	73.3	47
Methanol	61.4	39
Carbon tetrachloride	66.4	21
Chlorotrimethylsilane	57.0	7

Table 2 Vapor pressure of acrylonitrile over aqueous solutions at 273.15 K (Razdil et al. 2012)

Acrylonitrile (wt%)	Vapor pressure (kPa)
1	1.3
2	2.9
3	2.3
4	6.9
5	8.4
6	10.0
7	1.9

2.5.2 Application of Acrylonitrile

There are many end uses of acrylonitrile such as production of acrylic fiber, acrylonitrile-butadiene-styrene (ABS) resins, adiponitrile, acrylamide, carbon fiber and synthesis rubber name as nitrile rubber (Razdil et al. 2012). Synthesis rubber is introduced in Germany via base copolymer of butadiene and acrylonitrile. The addition of acrylonitrile makes the synthetic rubber is highly resistant to swelling in gasoline, oils and other non-polar solvents (Hansora 2014). Also, the synthetic rubber is applicable in the manufacture of gaskets, seals, hoses, belts and electrical cable jackets. During the Second World War, the production of synthetic rubber is increasing gradually due to lack of excess to natural rubber (Hansora 2014). According to Razdil et al. (2012), acrylic fiber is the highest use of acrylonitrile since the production demand is very high. Furthermore, acrylic polymers are used widely in textile applications (Gupta and Afshari 2009). Also, acrylonitrile is blending with fiber for home furnishing for example carpets and deep pile products such as fake furs and doll hair (Gupta and Afshari 2009). In the airline and automobile industry, carbon fiber is chosen for the application due to high strength and low weight. Figure 7 shows the percentage of end uses of acrylonitrile.

While, ABS and acrylamide are the fastest growing uses of acrylonitrile (Razdil et al. 2012). ABS is one of the high-performance polymer and easy to process (Razdil et al. 2012). Majorly, ABS is applied in numerous automotive, construction and electronic applications. For acrylamide, it usually found in the application for wastewater treatment, oil production, mineral processing and paper manufacture. Acrylamide is produced from acrylonitrile by the copper catalyzed process.

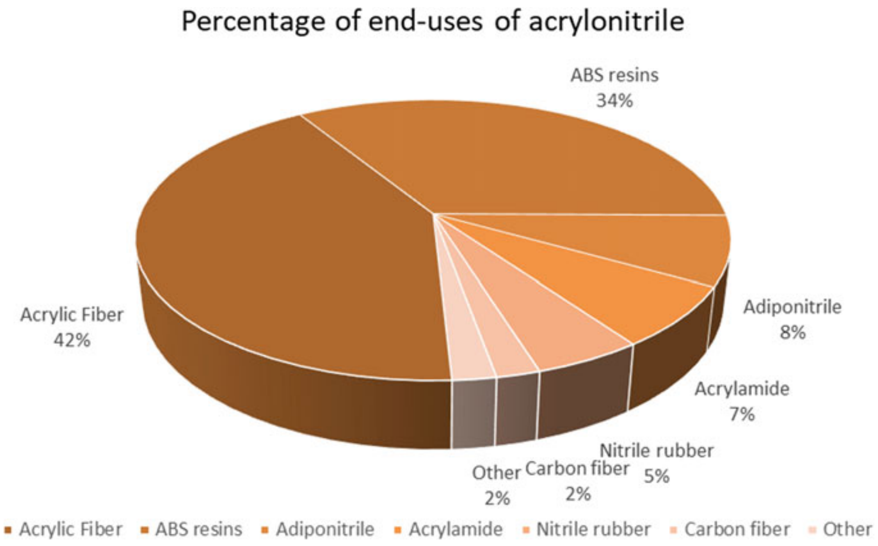


Fig. 7 Percentage of end uses of acrylonitrile (Razdil et al. 2012)

2.6 Enhancement of Wood Quality

Wood fiber cab extracted as a primary product or collected during the milling of lumber. Currently, the usage of wood fiber is growing perfectly due to their low cost and abundant in production. However, contain hydroxyl group and cavities exist in the wood cause the performance of wood decrease from the changes of dimensional (5–3.1). Due to existent of cavities on the wood component, it will increase the moisture of wood by providing the water to store in the cavities, thus hydroxyl groups contribute for wood fiber easily soluble in water (Rahman 2018). To overcome the problem, the wood should be treated by substituted the reactive site with some polymer or chemical. The polymer and chemical will fill the pores of the wood component thus make the wood more resistant towards water and decrease the moisture (Rahman 2018). Also, the addition of the reactive chemicals and monomer will increase the dimensional stability, physical and biodegradation properties of the wood. Means that the modified wood has a longer life and more sustainable in the application compare to the treated wood.

2.7 Modern Policies to Growth Wood Quality

Wood displayed abundant hydroxyl group and various cavities which led to some disadvantages features such as high-moisture uptake, biodegradation and dimensional instability (Rahman 2018). Due to unwanted features, the wood needs to be treated through various chemical treatments, single or dual impregnation to improve the quality of the wood (Rahman 2018). The main problem of the hydroxyl group and cell cavities contain in wood is they create the path for moisture movement in the wood component (Li et al. 2013; Rahman 2018). By introducing the modification process to the wood, the reactive site can be replaced with suitable polymer and chemical to block the pores and react with the hydroxyl group to change the properties of the wood (Rahman 2018). Also, the pores on the wood components could be blocked by the polymer by the impregnation process which enhanced the moisture resistant of the wood (Rahman 2018). In the study by Li et al. (2013), in situ polymerization reaction shows the best technique to reinforce the wood component with polymer with a different chemical reaction or monomer dispersion. However, the presence of hydroxyl group could make the wood hydrophobic to polymer matrix (Rahman 2018). So, the wood might hard to be enhanced.

There are two types of polymers that usually used in a modification which are thermosets polymer and thermoplastic polymers (Rahman 2018). Thermosets is polymers that have undergone a chemical reaction by the action of heat, catalyst, ultraviolet light and cannot be reworked and decomposed (Wegman and Van Twisk 2013). The thermosets that usually prepared for the reinforced composite are Bis-Maleimide (BMI), Epoxy (Epoxide), Phenolic (PF), Polyester (PL), Polyimide, Polyurethane (PUR), Silicone, Polyether ketones and Cyanate ester. Thermoplastic polymers are defined as the polymer that is foldable and flexible when the heat is applied to it. There are various types of thermoplastic polymer matrix used in composite materials such as Polybutylene (PB), Poly(vinyl) fluoride, Poly(vinyl) chloride (PVC), Poly(vinyl) acetate (PVAC) and Nylon (Rahman 2018).

Woods consists of two types which hardwood and softwood. According to Rahman (2018), hardwood is more expensive than the softwood because the durability of hardwood is higher that softwood. In past decades, most of the people and industry used hardwood for the application and end-use product instead of softwood because it's the simplest solution to create a product with high durability (Rahman 2018). However, the availability of the hardwood keeps decreasing because of the demand increasing in every year (Rahman 2018). To overcome this problem, researchers and manufacturer find the possible ways to increase the wood properties of the softwood by reinforced them with polymer and chemical. Four technique is basically employed to develop the wood form non-durable species of wood, which are chemical modification, impregnation modification, thermal modification and surface modification (Rahman 2018). In other research displayed, the main type of process should implement in the wood treatment are chemical treatment, thermo-hydro treatment (TH) and thermo-hydro-mechanical (THM) treatment, biological treatment and physical treatment by applying the electromagnetic irradiation or plasma (Sandberg et al. 2017).

Rowell (2006) states, chemical modification of wood is defined as a chemical reaction between the chemical component and the reactive site of the cell wall of raw wood. Usually, the formation of covalent bond existed between OH groups from cellulose, hemicellulose and lignin (Rahman 2018). The chemical modification of the wood will offer enhancement to the wood quality in terms of dimensional stability and biological resistance (Rowell 2006). The resistance of biological attack is probably due to low moisture content in the cell wall that needs for biological activity. They conclude the modified woods are more resistance from brown and white rot fungi because of the moisture reduction of the wood (Rowell 2006). While impregnation modification of the wood is defined as the cell wall of the wood is soaked into various type of monomer and polymer to fill the cell cavities and increase the biological resistance (Processes 2006; Rahman 2018). Thermal modification is conducted when the heat is applied to the wood components that result in degradation associated with chemical composition in the wood material (Rahman 2018). In the extensive research, proper thermal treatment changes the colour of wood, increase the hydrophobic properties, reduces the moisture content value and as well reduces the mass (Kondratyeva et al. 2016). Moreover, due to low moisture content, it will lowering the possibility of biological attack towards the wood (Kondratyeva et al. 2016). Next, the aim of surface modification is to change the wood surface by introducing the chemical and biological modification using enzymes and physical process such as plasma modification (Rahman 2018). In other words, it improves the bonding between wood fiber and the coatings material to ensure the wood can withstand in various type of weather and condition.

2.8 Chemical Modification of Wood

Wood is a natural product that originates from a different type of trees. Every type of wood has a limitation on the application and high-end product due to the toughness and strength of the wood component (Kondratyeva et al. 2016). So, modification technique is introduced to fix the weak points of the wood components that are mainly related to moisture sensitiveness, low dimensional stability, low hardness, biodeterioration against fungi and low resistance to ultraviolet (UV) radiation (Rowell 2006). Based on the review by Rowell (2006), he found the chemical modification of wood that displayed the consistent results mostly using acetylation process by using acetic anhydride as the primary polymer. Also, acetylation remained the most reaction chosen due to low cost and the global availability of acetic anhydride (Rowell 2006). Moreover, the durability of modified wood increases due to covalent bonding occurred during the acetylation process (Rahman 2018). The effectiveness of the acetylation process is increasing with time of reaction (Paril 2016). They conclude the rate of acetylation is increased with increase the power of the microwave which used to house the reaction between chemical or polymer and wood component (Paril 2016). This technique should be done in the vacuum condition to avoid the presence of moisture that brings by the air which may affect during the modification process. Effect of the acetylation reactions, the functionalization of the alcoholic function groups and natural hydrophilicity of wood is decreasing (Rahman 2018). Paril (2016) displayed, acetylation reaction changed free hydroxyl within the wood into acetyl groups.

2.9 Wood Modification by Impregnation Technique

Various types of chemicals or polymer are used to improve the wood properties by depending the moisture content, permeability and crystallizing point (Paril 2016). Wood impregnation is defined as a process to diffuse the gas or to inject the liquid into the wood component and the permeability of the wood cell (Rahman 2018). The wood component can be greatly enhanced by filling the empty pores and space with the various type of chemical and polymer (Rahman 2018). According to Li et al. (2013), there are many types of vinyl monomers and polymeric monomers have been used to prepared WPCs with improving properties such as styrene, vinyl acetate, acrylonitrile, methacrylate, acrylates and acrylamide. The raw sample or specimens basically should be free from air, moisture and water before promoting to impregnation process to ensure the pore of the wood is completely empty and easy for chemical or polymer to penetrate (Rahman 2018). Also, in another review, the preparation technique of reinforced resin is very significant to ensure the impregnation process is successful. Rahman (2018), states the treating chemical and polymer should be prepared with low viscosity, which normally corresponds to low or medium molecular weight. Coupling agents are defined as substances that are used in small quantities to treat a surface so chemical bonding betides between wood and polymers (Li et al. 2013). They add the coupling agent to overcome the problem when the polymer and resin do not fully graft with raw wood. This method is very useful for improving the affinity, compatibility and adhesion between wood cells and resin or polymer. There are various types of coupling agent used in the impregnation technique such as isocyanates, anhydride, silanes and epoxides. In some review states the most successful coupling agents are maleic anhydride (MAN) and glycidyl methacrylate (GMA) (Badji et al. 2016; Li et al. 2013).

2.9.1 Pre-treatments

Pre-treatment process is very significant before the wood undergo the impregnation process. The pre-treatment process is done to ensure the wood is well dry. Drying the wood will increase the preservative penetration and retention (Paril 2016). Means, the reinforced resin is easy to fill the pore and the holes of the wood component because already out of moisture. Also, prevent the microorganisms to growing on the wood component.

2.9.2 Non-pressure Impregnation

The impregnation treatment can be done in non-pressure condition. There are several techniques have been done for a non-pressure impregnation technique which is brushing and spraying, dipping, soaking, diffusion, and hot and cold batch (Paril 2016). A very simple and low-cost technique is brushing and spraying. However, the retention between polymer and wood is very limited. Also, the penetration of the chemical and polymer across the wood cell is minimal and some penetration along the grain is possible. The second method is dipping which it much better than brushing and spraying technique. By applying this technique, its increase end grain penetration and from the result found the pinewood able perforation depth in the range of 25–75 mm (Paril 2016).

Next technique is known as soaking treatment. Soaking is similar to the dipping technique but difference of the immersion time, which could be hours, days or weeks (Paril 2016). Basically, this treatment is applying by the sawmill for small poles and lumbers to reinforce the raw wood used for the production. The other technique is known as diffusion treatment which usually applied to freshly sawn green wood with moisture content in excess of 50% (Paril 2016). A waterborne preservative such as boron salts, fluorine compound is prepared in paste form. Thus, the surface is the wood is brushed with the waterborne preservative. During the diffusion process, the solution will flow from higher concentration to lower concentration area. So the concentration gradient across the material should take place to ensure the properties of impermeable species is improved.

2.10 Presence of Clay in Wood Impregnation

Clays minerals are belonging to a main group of silicates with a layered structure known as layered silicates and mostly used in polymer nanocomposite preparations due availability and have low cost (Olad 1996). Modification of wood in the presence of clays has been proven to be very effective in improving the various properties of wood (Rahman 2018). In the extensive research, the researcher undergoes the modification of wood through in situ polymerization with the addition of silicate and nanoclays. The clay presence act as compatibilizer that accomplishes the motion of polymer chain in the modification of acrylonitrile/butyl methacrylate/halloysite nanoclays wood polymer Nanocomposites (AN-co-BMA-HNC WPNCs) (Rahman 2018).

There are many improvements in wood properties with the addition of the clay. Based on the study by Rahman et al. (2013), they conclude the thermal stability of WPCs is much better than the control wood due to the coupling reaction with cellulose and new chemical bonding between styrene and methyl methacrylate with the aid of benzoyl peroxide and nanoclay. In the study conducted by Devi and Maji (2013), they displayed the thermal decomposition of modified wood inclusion of nanoclay is higher than the raw wood which means the temperature for wood to decompose is increases. The high value of decomposition temperature is due to the crosslinking created by glycidyl methacrylate (GMA) whereby reduce the rate of degradation of degradable components like carbon monoxide (CO) and carbon dioxide (CO_2) (Devi and Maji 2013). The positive result is displayed by Devi and Maji (2013), which are the silicate layered act as a barrier to inhibit the diffusion of volatile decomposition products.

According to the research of Rahman (2018), they conducted modification wood by impregnated wood with melamine-urea-formaldehyde (MUF) resin and wood impregnated with MUF resin/nanofiller to have the comparison on the sample specimen on water absorption and dimensional stability. Three types of specimen with different additives are immersed in the water to identify the most water uptake of all wood specimen. Table 3 describes the result of the water absorption by each specimen done by Rahman (2018).

From the result in Table 3, its clearly shows the performance of wood impregnated with MUF resin and organophilic nanoclay has lesser water absorption than the wood impregnated with MUF resin only and the control wood. Also, due to low

Table 3 Percentage of water absorption of each specimen (Rahman 2018)

Type of specimen	Water absorption (%)	
	After 24 h	After 1 week
Control wood	63	125
MUF resin-impregnated wood	8.3	38.5
Organophilic nanoclay/MUF resin-impregnated wood	5	22

water moisture content, it also has high resistance of biological attack among the three types of specimens. The presence of clay will help to improve mechanical performance properties and reduce the water absorption as well (Rahman 2018). Besides that, according to Rahman (2018), the addition of nanoclay is significant to improve the mechanical properties of WPNCs especially the modulus of elasticity (MOE).

2.11 Wood Deterioration

There are many reasons for the deterioration of wood components. The wood deterioration can be divided into two types of factors which are abiotic factors and abiotic factors.

2.11.1 Abiotic Factors

The biotic factor is defined as a non-living factor in an ecosystem. As part of the ecosystem, these factors existing around and affect the living things in it. Common examples of abiotic factors are wind, rain, humidity, temperature, soil composition, pollution, ultraviolet (UV) radiation and salination (Howell and Baynes 2007; Kirker and Winandy 2014). There are three modes of abiotic ways which are weathering, anatomical and mechanical action (Kirker and Winandy 2014; Umachandran and Sawicka 2017). The surface of the wood will be changed when woods are exposed to the outdoor and direct effects of the rain, freeze-thaw and wind. The penetration of ultraviolet (UV) of sunlight will cause wood degradation by the deterioration of the lignin (Umachandran and Sawicka 2017). As deterioration and reduction of lignin have occurred, eventually it will reduce the bonding of the wood fiber. Thus, it led to the degradation by physical and mechanical properties of the wood and allow the wood to promote UV light penetrates deeper. Other than that, a biological attack is one of the factors led to wood degradation (Umachandran and Sawicka 2017). In the extensive research, presence of the microorganism causes the changes of the natural color of the wood due to dark-colored fungal spores and mycelia on the wood (Howell and Baynes 2007; Umachandran and Sawicka 2017).

2.11.2 Biotic Factors

Damage of the wood of abiotic factor is referred to the damage done by the organism which include fungi and insects (Umachandran and Sawicka 2017). The growing of the fungi in wood is basically the due presence of moisture. Umachandran and Sawicka (2017) state, moisture management is the most critical work to handle in wood management. The multicellular microorganisms are growing very fast in the component of the wood, so it increases the rate of wood decay. There is some type of fungi that disturbed the structural of the wood such as brown-rot fungi, white-rot fungi, soft-rot fungi, mold fungi and sap stain fungi (Kirker and Winandy 2014; Umachandran and Sawicka 2017). Moreover, the most famous insect that always causes the wood damaged is termites. It is because wood is the main source of food and one of the shelters for termites. Severe termite damage can cause disastrous wood failures, especially in the internal damage of the wood (Kirker and Winandy 2014).

2.12 Wood Timber and Wood Impregnation Industry and Market

The forest products industry is an important socioeconomic sector to many developing countries especially in terms of foreign exchange, earnings and employment. Malaysia is well known as developing countries. The transformation from an agricultural based economy to manufacturing based company over half a century in Malaysia has been remarkable (Ratnasingam et al. 2013). Figure 8 shows the trends of agricultural and manufacturing sectors to the gross domestic (GDP) of Malaysia.

The richness of natural resources and enough supply of power source has led the industry has grown rapidly with an annual growth rate of 10% since the 1980s. However, from 2005 to 2010 graph shows a downtrend in manufacturing economy-based forest resource due to the presence of competitor that offers low-cost product especially China and Vietnam (Ratnasingam et al. 2013). The annual growth rate of subsectors in the forest products industry has been inconsistent, although the furniture and plywood sectors appear to be more positive. This was probably due to the relatively higher level of innovation in these subsectors (Ratnasingam et al. 2013).

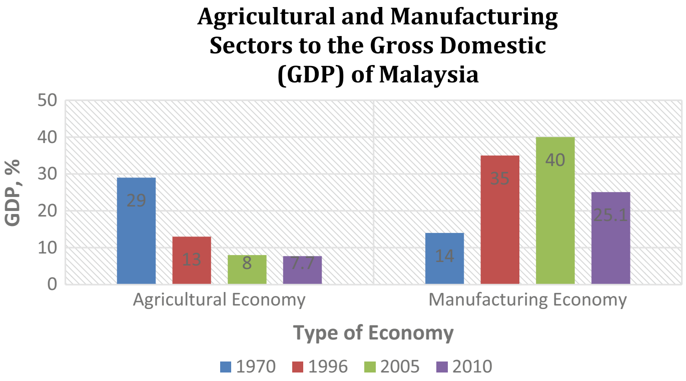


Fig. 8 The trends of agricultural and manufacturing sectors to the gross domestic (GDP) of Malaysia (Ratnasingam et al. 2013)

The wood furniture industry has become the largest subsection accounting for nearly one-third of the country's total export receipts. Under the industrial master plans, the industry was designated as a target industry and special incentives were given to boost export growth. Eventually, it is no surprise that the industry's ratio of export/production is 80% (Ratnasingam et al. 2013). Moreover, Ratnasingam et al. (2013) states, factors like marketing communication, technology, cost of production and demand have a strong effect on the comparative advantage of Malaysian furniture. Despite Malaysia's encouraging growth and export performance in the forest products sector, it is clear that the industry is lagging behind innovation and creativity compared to its counterparts in Germany and Italy (Ratnasingam et al. 2013). The need for innovation and creativity in forest products is a necessity for the industry of sustainable forest products worldwide. Wood impregnation industry and market are keep growing around the world. According to Paril (2016), 6.5 m^3 of pressure treated wood is produced by preserving industry in Europe for every year. Wood modification is a very significant method as a production step in sawmills or other wood processing industries. Figure 9 shows market share categorized by impregnate and products.

3 Methodology

3.1 Materials

The species of wood used for this study was acacia wood. Acacia wood was obtained from the Forestry Department Sarawak, Malaysia. The poly(vinyl) alcohol matrix used in this project was in powder from with density of 1.38 g/cm^3 and it was supplied by Sigma-Aldrich Co. The type of clays was involved in the preparation was manufactured by BYK additives Inc. the solvent used was acrylonitrile which obtain form Sigma-Aldrich Co.

3.2 Equipment and Instruments

The equipment used in the research project were Autograph Mechanical testing apparatus from Shimadzu Corporation from Shimadzu Corporation, sonicator (Elmasonic S900H), SEM (Hitachi TM3030 Tabletop), Fourier Transform Infrared Spectrophotometer (IRAFFINITY-1 CE Shimadzu). The laboratory apparatus used were beakers, spatulas, glass rods, electronic balance and mechanical stirrer. The impregnation wood process was done in the vacuum chamber in the fume hood.

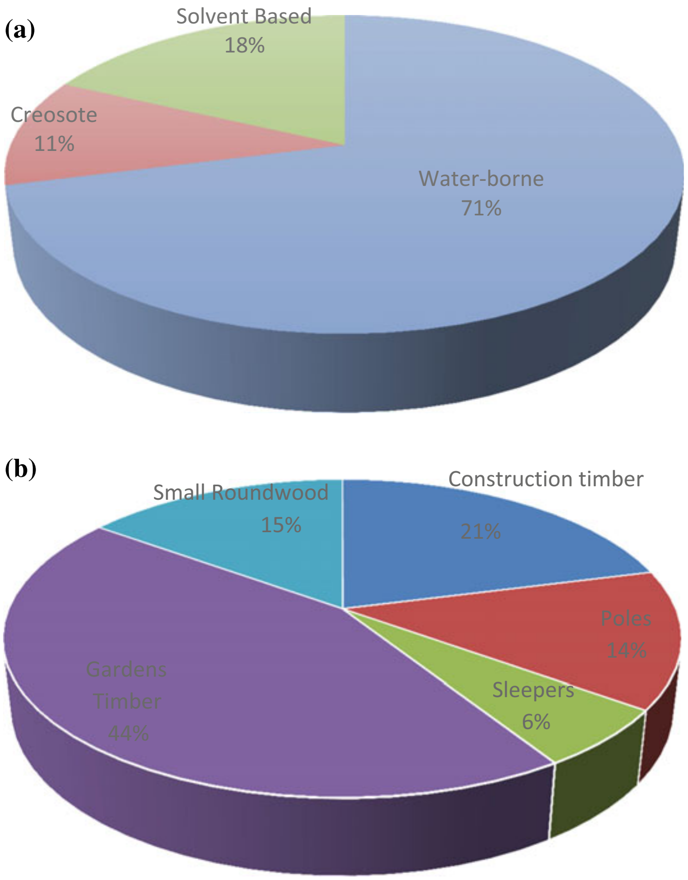


Fig. 9 European wood preserving industry production categorized by **a** impregnate type and **b** by type product

3.3 Impregnated Wood Preparation

The wood samples were prepared in the dimension of 240 mm length × 20 mm width × 10 mm thickness. A total of 27 wood samples were prepared with the same dimension and 3 wood samples were used for each treatment combinations. A set of untreated wood with 3 samples were served as control. The resin solutions were prepared in this study, which are mixture of poly(vinyl) alcohol (PVA) and clay with the solvent of acrylonitrile which is 200 mL. A set of experimental design of the study is shows in Table 4. All the treated wood samples were impregnated in resin solution using vacuum pressure process with the desired of time. The design of the experiment was prepared by using Design Expert Software.

Each of the wood samples were weight before and after the modification of the wood polymer nanocomposites (WPCS). The weight percent gain (WPG) of all

Table 4 Experimental design of the study

Name of sample	Treatment method	Concentration of PVA (%)	Weight of clay (g)	Time (min)
UT1	Untreated (control)	–	–	–
PVA1/10C/10M	Impregnation	1	12	60
PVA8/1C/10M	Impregnation	1	2	60
PVA1/1C/60M	Impregnation	8	12	10
PVA1/10G/60M	Impregnation	1	12	10
PVA8/1G/60M	Impregnation	8	2	10
PVA1/1G/10M	Impregnation	8	12	60
PVA8/10G/10M	Impregnation	1	2	10
PVA8/10G/60M	Impregnation	8	2	60

the wood samples were then measured using equation below. After that, the wood samples were wrapped with aluminum foil and kept in an incubated oven at 100 °C for 48 h for polymerization to take place.

$$WPG = \frac{W_f - W_o}{W_o} \times 100\%$$

where,

W_f over-dried weight after modification of the WPNCs
W_o over-dried weight before modification of the WPNCs.

3.4 *Characterizations of Impregnation Woods*

Morphological, mechanical properties, chemical structures and surface area investigation of modify and control acacia wood were studied by the characterization study and analysis of the samples prepared. The mechanical properties were studied by the universal testing machine. The Scanning Electron Microscopy (SEM) studied the morphology and the Brunauer-Emmett-Teller (BET) studied the specific area of the samples. Fourier Transform Infrared Spectroscopy was used to study the chemical structure of acacia wood.

3.4.1 Mechanical Properties (Tensile Testing)

The study of mechanical properties of Acacia woods were determined and evaluated by tensile properties. Tensile strength and Young's modules were found and comparison study was done according to the weight of copolymer and clays added with different time taken for impregnation process. The tests were conducted with Shimadzu Universal Testing machine that had a loading capacity of 300 kN and cross head speed for the test was 1 mm/min. The test was run to detect the force that the prepared samples could withstand. Tensile test was conducted for all samples. The wood specimens were tested accordance to ASTM D638 and analyzed at room temperature with stretch rate of 1 mm/min. the results of tensile strength for each wood specimens were displayed by the computer that connected to the machine. The testing was running by using Trapezium 2 software.

3.4.2 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier Transform Infrared Spectroscopy (FTIR) is one of the most popular analytical analyses that commonly used in various field such as pharmaceutical, life science and electron industries. This characterization included structural information of materials, analysis of materials as well as determination of contaminants IR Absorptions of Functional Group (22, flash Exchange). This equipment was used to determine the chemical change upon impregnation of the raw Acacia wood with impregnation of chemical substances.

3.4.3 Scanning Electron Microscope (SEM)

The morphology of the wood samples was studied by a tabletop scanning electron microscope. The interfacial bonding between the acrylonitrile, poly(vinyl) alcohol and nanoclay were examined using a SEM. The SEM analysis was used to detect the pore size of the treated and untreated wood samples. The primary electron beam will scan across the surface of the wood samples. Thus, the signal will generate when the electrons strikes the wood samples. After that, the specific signals will produce an image of the surface of wood samples. The wood specimens were first fixed with Karnovsky's fixative. Thus, wood samples were cut into the desired dimension before being viewed microscopically.

3.4.4 Thermogravimetric Analysis (TGA)

Thermogravimetric Analysis (TGA) is carried out to study the thermal analysis of treated and untreated wood samples. This characterization is used to determine the degradation temperatures of absorbed moisture content of materials especially on wood polymers. The process involved heating a mixture to a high enough temperature to decompose the samples into a gas. The TGA testing was started by placed the samples on sensitive balance that measure the weight loss of the sample as the system was heated. It was identifying the behavior of weight percentage against temperature. TGA analysis was used to investigate the corrosion kinetics in high-temperature oxidation. After the data were obtained, curve smoothing, and other operations could be carried out to find the exacts point of inflection. In this method, temperature increased affect the increased of weight loss.

3.4.5 Differential Scanning Calorimetry Analysis

Differential Scanning Calorimetry (DSC) is a technique used to study the thermal transitions of a substance when it is being heated. Thermal transitions are the changes that take place in a polymer such as the melting of the crystalline and the glass transition of the substance. In this work Q 1000 DSC system (TA Instruments, USA) was used to determine the influence of addition of polymer and clay loading on the crystallization and melting behavior of the wood plastic composites. Each sample was heated and cooled at a scanning rate of 10 °C/min under nitrogen atmosphere in order to prevent oxidation. A test sample of 5–10 mg was placed in an aluminum capsule and heated from 20 to 600 °C for each run.

4 Results and Discussion

4.1 Mechanical Properties (Three Points Structural Testing)

The Three-Point Structural Testing of treated wood and untreated wood is to determine the amount of resistance to bending while applying the vertical force to the sample. Flexural strength is calculated by referring to the strength of wood to resist load applied until it reaches the breaking point. The table shows that the treated wood specimens exhibited higher flexural strength value than the untreated wood specimens. This was due to the presence of fillers which are PVA and nanoclay that exerted Van der Waals forces within the wood cell wall (Hamdan and Mazlan 2018). Additives of polymer and nanoclay were found to enhance the mechanical properties of the wood. Furthermore, the vacuum time of the wood impregnation affect the changes in the treated wood properties. It has been observed that the use of a "vacuum break" enhances chemical penetration into wood, and they are an essential element in the basic impregnation process. The vacuum break is the end of the vacuum period when the impregnant is introduced and permitted to penetrate the wood. Figure 10 shows the bar chart of the flexural strength for the untreated sample and treated samples.

Among the treated wood specimens, the highest value of flexural strength was from the sample PVA1/10C/10M which contain 1% of PVA and 10 g of nanoclay with 10 min of vacuum time which time consume for wood impregnation with the value of 118.273 N/mm^2. While the lowest value of MOR of the treated wood specimens was from the PVA1/10C/60M with the value of 83.0428 N/mm^2. The evidently improved mechanical properties of the treated wood were attributed to the formation of PVA with excellent performance and the good complex effect of the resulting polymers

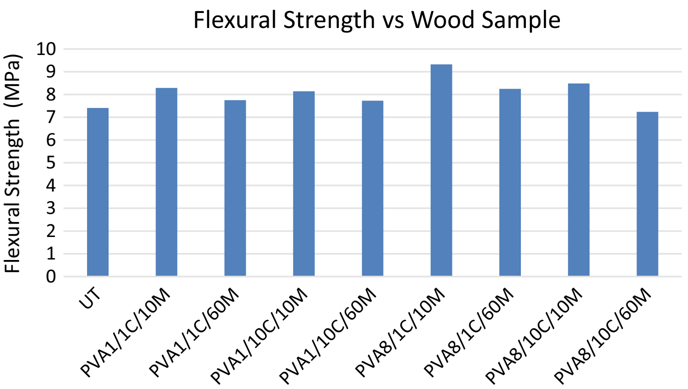


Fig. 10 Flexural strength of untreated wood and treated wood with different value of PVA, nanoclay and vacuum time

with the wood cell walls. Furthermore, the increment of the percentage for the MOR increases to 42% for the wood specimen of PVA1/10C/10M compare to untreated wood. Means that, the additives of the nanofiller gives major improvement for the strength of the woods. Moreover, satisfactory compounding of the resulting PVA and nanoclay with the wood cell walls allowed the load transfer from the wood to the polymers. Thus, the part of forces passing on to the polymers, which also contributed to the improvement in the mechanical properties. Table 5 illustrated the summary of all values of each sample.

The comparison between the wood sample of PVA1/10C/60M and PVA1/10C/10M shows the lowest and highest increment of the MOR value which are 3.03 and 42.42% respectively. The value of additives of nanofiller for both samples was similar which is 1% concentration of PVA and 10 g of nanoclays. However, there were differences in the vacuum time that makes the value MOR for both samples is the difference. Also, similar cases were shown by the data between sample PVA1/1C/10M and PVA1/1C/60M, sample PVA8/1C/10M and PVA8/1C/60M,

Table 5 Summary of mechanical properties for treated wood and untreated wood

Sample	Flexural strength (MPa)
UT	83.0428
PVA1/1C/10M	110.724
PVA1/1C/60M	95.625
PVA1/10C/10M	118.273
PVA1/10C/60M	85.5592
PVA8/1C/10M	115.757
PVA8/1C/60M	88.0757
PVA8/10C/10M	115.757
PVA8/10C/60M	98.1414

sample PVA8/10C/10M and PVA8/10C/60M. From the result can be concluded, the optimum time for impregnation will give the higher the value of MOR of the treated wood. Similar observations were prepared by Rodrigues et al. (2004) while exploring the compressive strength of impregnated Eucalyptus Grandis wood with different of vacuum time for wood impregnation. The shorter processing time of wood impregnation such as that would yield significant savings and added value to the modification wood industry. Moreover, the retention time of chemical to penetrate the permeability of the wood component could be concluded. Whereas optimum time is needed to ensure PVA and nanoclay creates a chemical bonding between wood structure. Means that, the increased value of MOR have vary trend with the extending of impregnation time. The stress–strain curve for raw wood and impregnated wood were showed in Fig. 11.

The variations of tensile strength were shown in Fig. 11. Clearly shows, the tensile strength of the treated wood samples were higher than the raw wood sample. This is due to strong interfacial adhesion of chemical and nanoclay with the wood. Also, the void contents in wood component were excellently filled with nanofiller. According to Rahman (2018), the good impregnation of the wood could reduce the formation of void on the wood surface that enhanced the stress transfer between the polymer matrix and fiber. So, with the reduction of void formation, it would improve the cross-section area of the wood to interact polymer matrix effectively.

Figure 12 shows the stress–strain curve of the raw wood and treated wood sample with the formulation of 8% of PVA, 10 g of clay and 60 min of impregnation time. It can be observed from the curve, stress–strain curve of the untreated wood showed a linear elastic region at the beginning of compression. Thus, followed by a yield point,

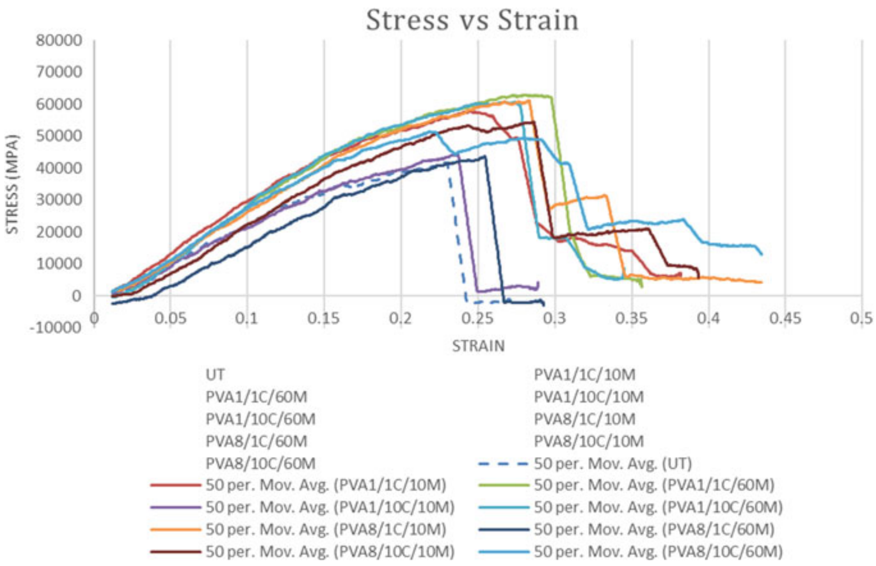


Fig. 11 Stress–strain diagram value of raw wood sample and each treated wood sample

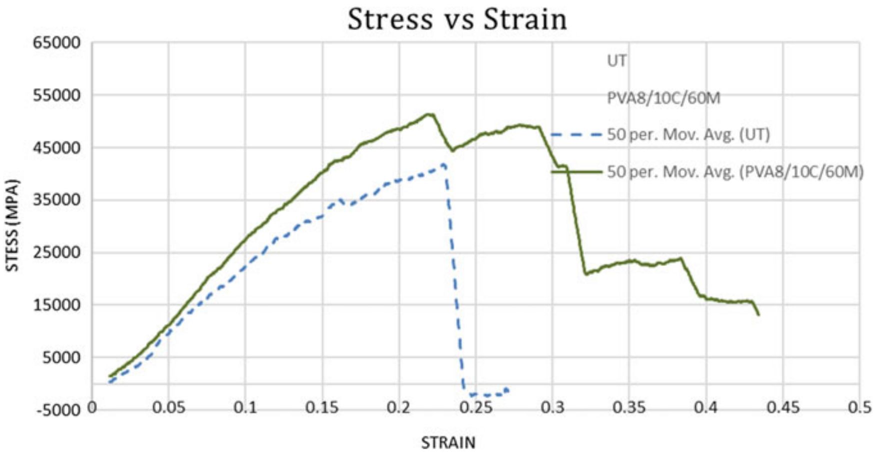


Fig. 12 Stress versus strain curve of untreated and treated acacia wood PVA8/10C/60M

indicating the onset of cell wall buckling and collapse. Then, the wood continued to deform under compression with a drastically decreasing stress. Meanwhile, the treated wood samples showed slightly similar trend at the beginning compression. However, higher Young's modulus indicated by the slope of the linear portion of the stress–strain curve. Furthermore, elevated stress threshold required to initiate cell wall collapse was observed for the treated wood. After the yield point, the treated wood showed an initially abrupt and the steady decrease in compressive compare to the untreated wood. Form this observation, indicates that the wood stiffness and yield strength in compression were improved by modification of the polymer and nanoclay.

4.2 Chemical Structural Analysis (FTIR)

FTIR characterization provides information about the chemical structure of samples of wood. Fourier transform infrared spectroscopic measurement of untreated wood and treated wood specimens were analyzed and recorded by Shimadzu Fourier Transform Infrared Spectrophotometer (IRAFFINITY-1 CE) at 27 °C, with a spectra resolution in range of 400–4000 cm^{-1} were measured to evaluate the potential interactions between Acacia Wood with PVA and nanoclays. These assignments were based on the values provided by the literature review (Kondratyeva et al. 2016).

It can be seen in Fig. 13, acacia wood contains nanoclay showing peak arising between the wavelength of 1000 and 1110 cm^{-1}. This is due to silica oxide (Si–O) stretching (Yadav and Yusoh 2016). The highest peak of the treated wood was from samples PVA1/10C/10M. Means that, the interaction of silicate and the wood component was very excellent compared to other treated woods. Also, from Fig. 20, the

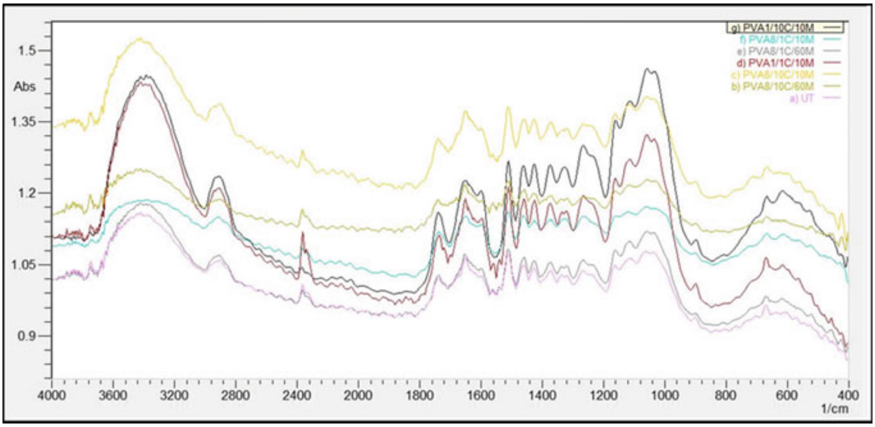


Fig. 13 FTIR spectra of seven types of wood samples

curve of samples PVA8/1C/60M shows the lowest value of absorbance and peaks due to silicon oxide stretching, among the others treated wood sample. Furthermore, the formation of these peaks were the indications of the presence of Si–O bonds due to the incorporation of the silicates of nanoclay in the acacia wood and this observation is similar with literature (Frankowski et al. 2007). Since silica was the dominant constituent of nanoclay whereby it is present in the tetrahedral layer of the clay. So, from the FTIR analysis is can be confirmed that the layered silicates (Si–O) were able to disperse and fill the void on the wood component. Also, the chemical bonding between PVA into the wood cell would certainly enhance the matrix adhesion and contribute to its property enhancement.

The region of spectra in between 3300 and 3500 cm^{-1} was used to identify and characterize the hydrogen bonded (O–H) and O–H stretch in the spectrum of the untreated and treated Acacia wood. The peak intensities of hydroxyl group wood and wood composites were produced by wood (Hamdan and Mazlan 2018). Meanwhile, the characteristics of broad peak that were observed at 2800–3000 cm^{-1} are attributed to the symmetric and asymmetric C–H stretching in aromatic methoxy groups. Moreover, the absorption band between 1600 and 1730 cm^{-1} indicated the stretching vibration of the carbonyl group (–C=O) caused by intermolecular H-bonding. The results also agree with the previous publication of Viet and Tho (2017) who reported the band is assigned for aromatic skeletal vibration.

A decreasing in the intensity of the transmittance of the O–H absorption band in Figs. 15, 17, 19, 20, 21, 22 around 3434 cm^{-1} explained that the hydroxyl group content in modified wood was reduced after finishing the treatment by PVA and nanoclay impregnation. The peak in range 3300–3500 cm^{-1} of wavelength was assigned to the stretching vibrations of hydroxyl groups for all samples. The peak at 3445 cm^{-1} was assigned for UT samples was shown in Fig. 14. This peak became prominently weaker than that of sample PVA8/1C/60M and slightly weaker than that of PVA8/1C/10M. Thus, the weakest peak was from the samples PVA8/10/10C. From

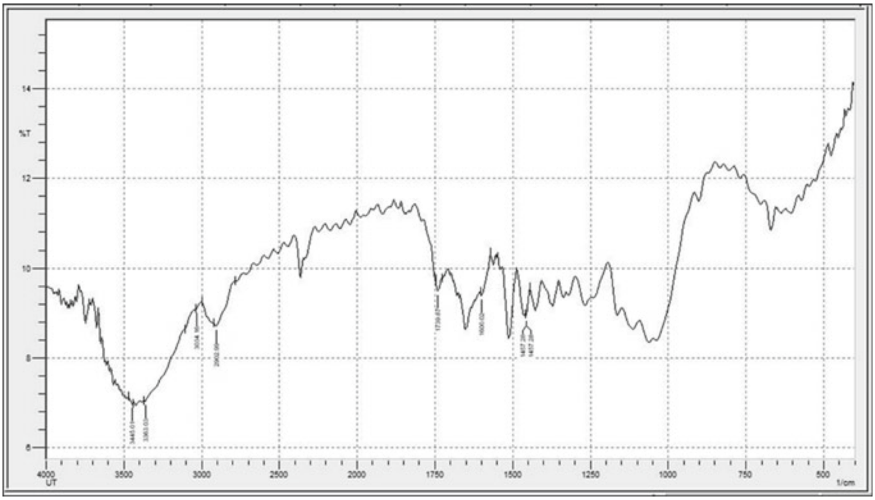


Fig. 14 IR spectra of untreated acacia wood

these observations, can be suggested that the relative number of hydroxyl groups in wood decreased due changed of some degree after the reaction of hydroxyl groups on the wood cells with PVA. According to Li et al. (2013), decrease in hydroxyl groups was likely due to the nucleophilic substitution reaction of a large number of hydroxyl groups on the wood cell walls with the polymer. So, the grafting of the PVA polymers on the wood cell walls resulted in a decrease of the hydroxyl numbers in the whole wood. Furthermore, it can be observed from Fig. 14, where UT sample has the highest number of wavelength compare to the lowest value from the PVA8/1C/60M sample that showed in Fig. 20 with value 3363 and 3343 cm^{-1} respectively. The corresponding peak shift to lower wavelength numbers was probably due to the transfer of some hydroxyl groups from the wood cell walls to the polymer chains.

Hygroscopicity was highly correlated with the accessible hydroxyl groups in wood. Since the main goal of modification is to improve dimensional stability and durability, the reduced hygroscopicity of the wood after treatment is an important parameter. However, there was two curves that showing the peaks of the hydroxyl group were higher than the peak of the untreated sample which is PVA1/1C/60M sample and PVA1/10C/60M sample. Means that, these two treated samples had contained high of hygroscopicity than the UT sample (Figs. 15, 16, 17, 18, 19, 20, 21 and 22).

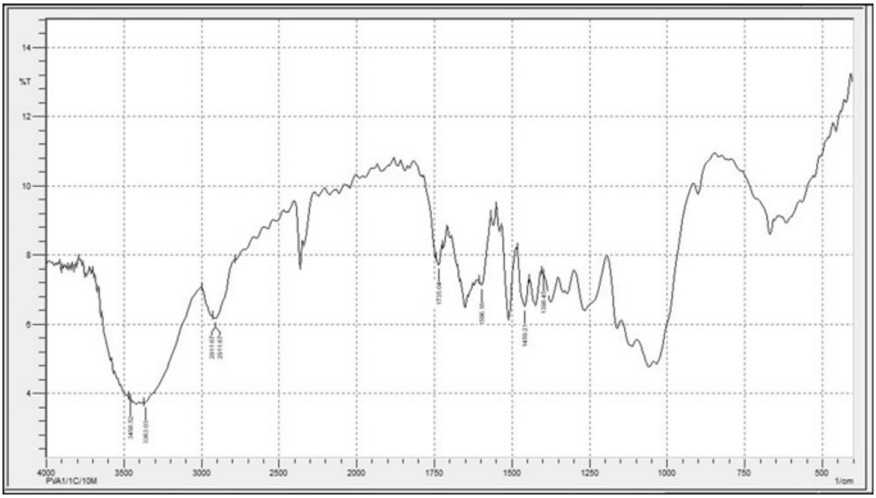


Fig. 15 IR spectra of treated acacia wood PVA1/1C/10M

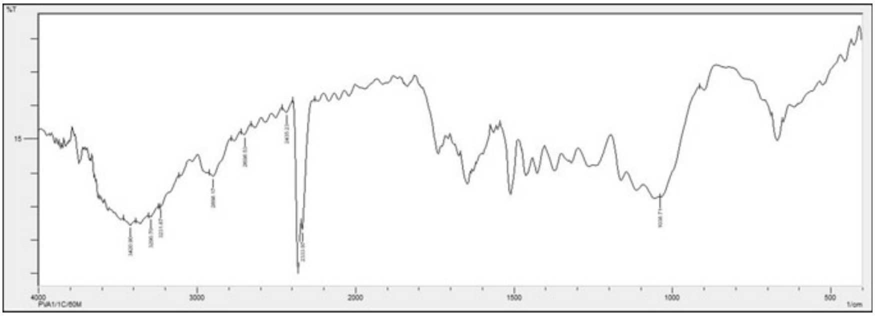


Fig. 16 IR spectra of treated acacia wood PVA1/1C/60M

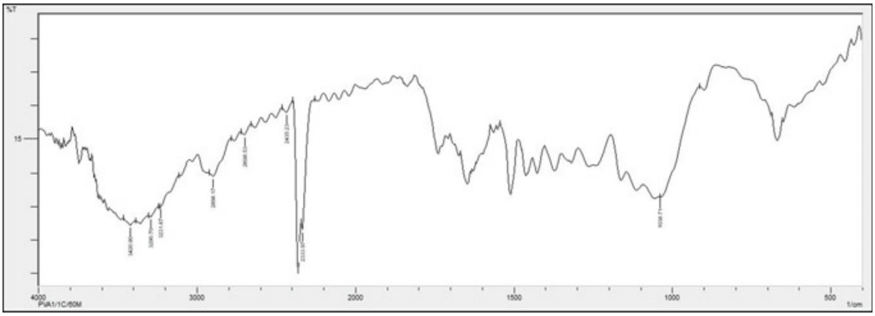


Fig. 17 IR spectra of treated acacia wood PVA1/10C/10M

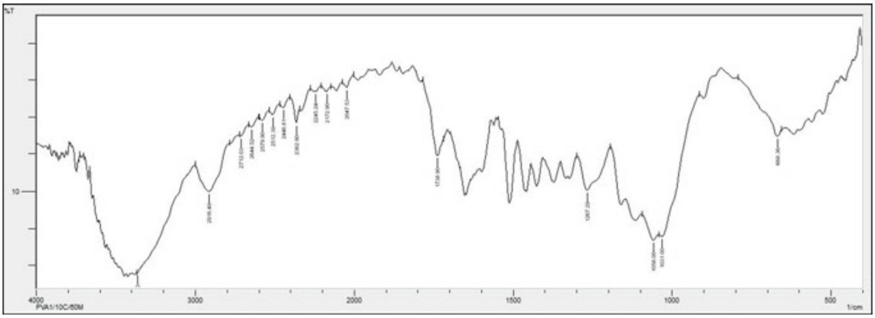


Fig. 18 IR spectra of treated acacia wood PVA1/10C/60M

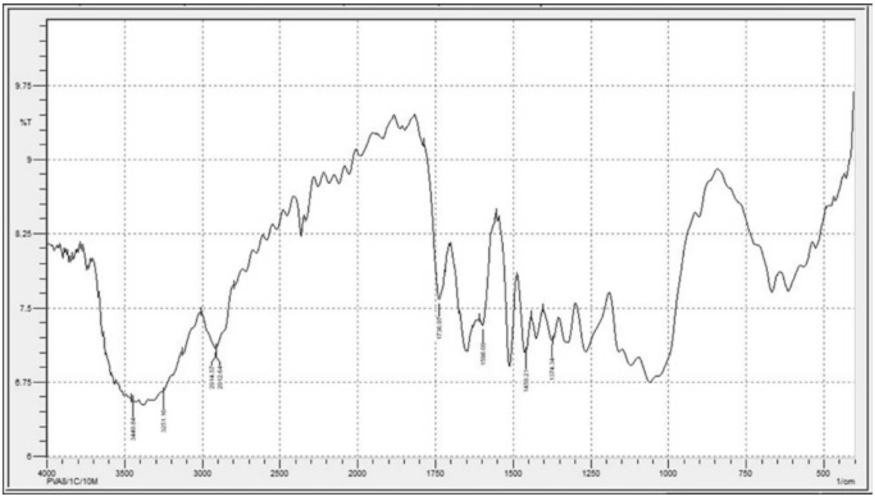


Fig. 19 IR spectra of treated acacia wood PVA8/1C/10M

4.3 Morphological Study (SEM)

The photos of sample surfaces, subjected to impregnation by the described methods were taken with the use of microscope SEM. The figure indicates that there appear significantly different in the interfacial interaction between fiber, nanoclay and polymer matrix in the composite system. Obviously, can be seen that the surface of untreated wood is rougher than the treated wood due to the chemical modification as well as polymerization that enhanced the hydrophobicity of wood fiber (Hossen et al. 2018). Whereby, the moisture of the wood has been replaced with the PVA and nanoclay (Figs. 23 and 24).

It was observed that the pores and porous of the wood were filled with the PVA polymer and nanoclay that make the differences from the untreated wood sample.

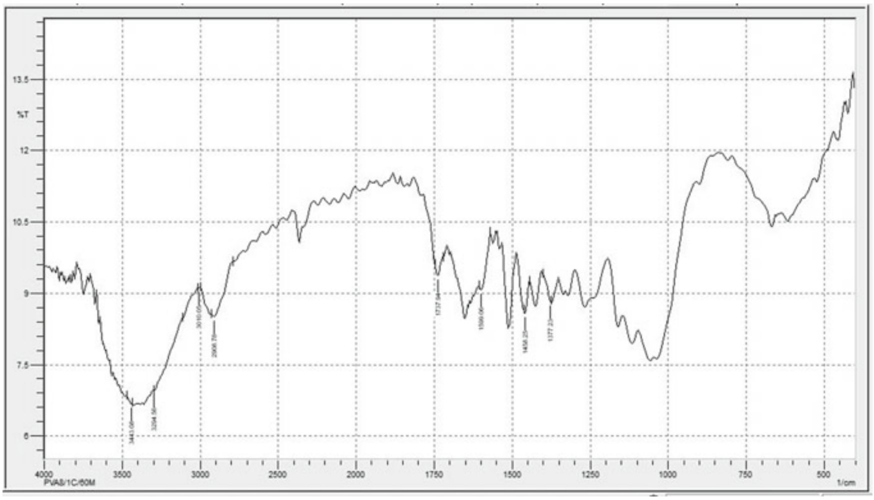


Fig. 20 IR spectra of treated acacia wood PVA8/1C/60M

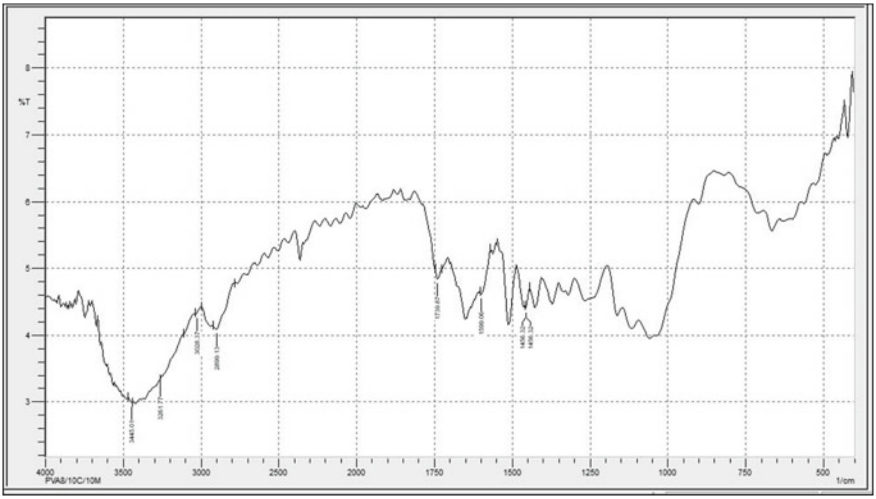


Fig. 21 IR spectra of treated acacia wood PVA8/10C/10M

The surface of Fig. 23c was smoother than those shown in Figs. 23a, b, d and 24a–e. In addition, the clay-incorporated wood polymer composites (WPC) showed less fiber pull out from the fractured surface and better dispersion of filler Fig. 23c. Moreover, SEM images from Fig. 23b and d, shows most rough surfaces and indicate the poor interfacial bonding between PVA, nanoclay with the wood component. Whereby, there were much different of the SEM images of PVA1/10C/10M sample and UT

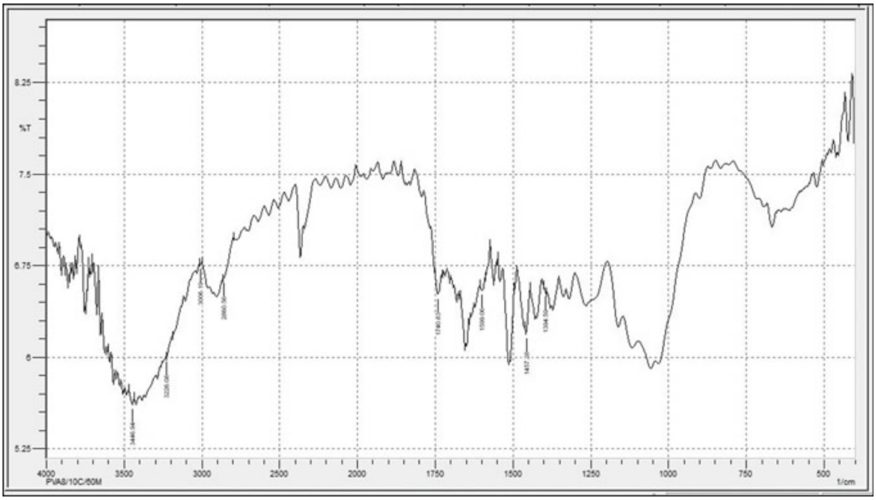


Fig. 22 IR spectra of treated acacia wood PVA8/10C/60M

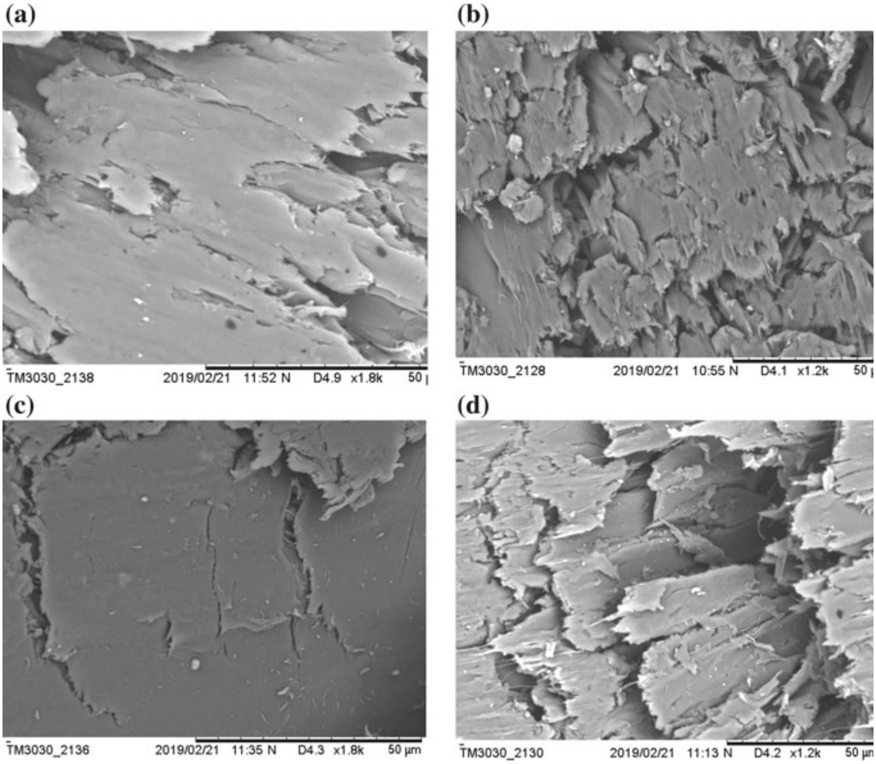


Fig. 23 Scanning electron micrograph of **a** PVA1/1C/10M, **b** PVA1/1C/60M, **c** PVA1/10C/10M, **d** PVA1/10C/60M

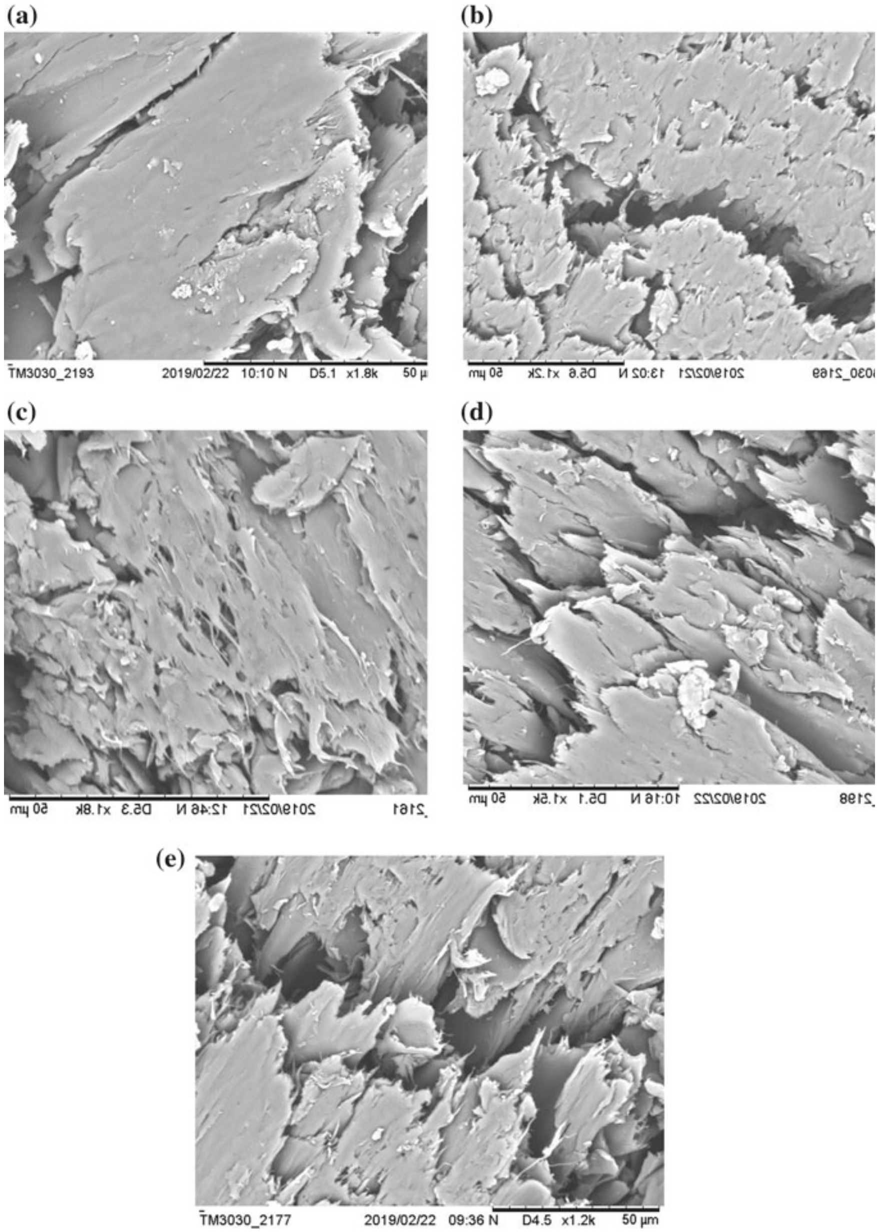


Fig. 24 Scanning electron micrograph of **a** PVA8/1C/10M, **b** PVA8/1C/60M, **c** PVA8/10C/10M, **d** PVA8/10C/60M, **e** UT

wood sample which means the wood component was effectively reinforced with the polymer and nanoclay within the desired of vacuum time.

Besides, the consumption of the vacuum time was very important to ensure the porous of the wood filled with the polymer and nanoclay before the vacuum break. The SEM image of PVA1/10C/10M in Fig. 23c and PVA1/10C/60M in Fig. 23d, shows a number of agglomerations of PVA and nanoclay in the wood component. This feature concluded that although amount of nanoclay and concentration of PVA were similar for preparation of both samples, the less time is sufficient to introduce nanofiller to penetrate the wood. From observations of Fig. 23b and d, found that PVA, as a single polymer, did not chemically react well with the wood cell walls during polymerization in the extending time. Degree of dispersion of nanoclays and polymer in the wood cell also the nature and intensity of filler-polymer adhesion interaction greatly affect the performance in mechanical properties. Also, the strong interaction of polymer and nanoclay between wood component could have resulted in the sample of PVA1/10C/10M.

4.4 Thermogravimetric Analysis (TGA)

The thermal stability of wood is a very important parameter in the production of modified wood with chemical and nanoclay additives. PVA and nanoclay are used as filler material to improve mechanical strength and other properties. For thermal properties analysis, Thermogravimetric Analysis (TGA) is carried out where the change in weight of the sample was recorded as a function of time with temperature ranging from 30 to 600 °C. This characterization is used to determine the degradation temperatures or absorbed moisture content of materials, especially on modified wood.

The thermogravimetric (TG) curves for the raw wood and modified wood are shown in Fig. 25. The mass loss process of all samples could be divided into three stages such as water evaporation phase, thermal degradation phase and carbonization

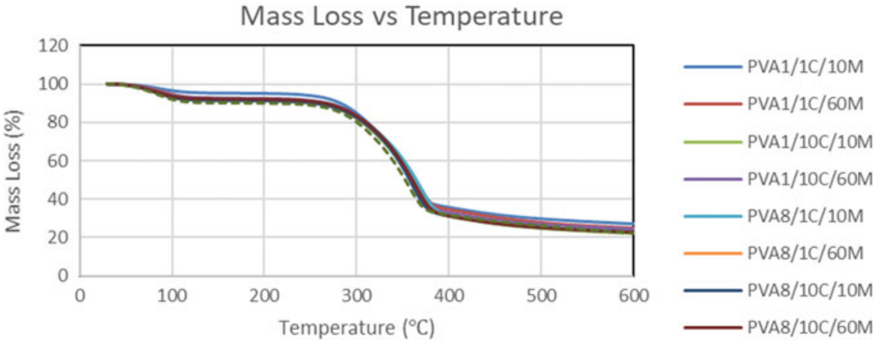


Fig. 25 TGA thermogram of untreated wood and modified wood

phase according to the rate of mass loss (Liu et al. 2018). The curves for the control and modified wood samples showed nearly similar trends however there are different in the percentage of mass losses for every temperature. From the TGA curve in Fig. 25 can be conclude, Acacia wood decomposed in the temperature range of 100–600 °C with three distinct weight loss stages.

Initially, the thermal degradation of the samples shows a slow decrease in weight loss below 220 °C. The initial weight loss from room temperature to 220 °C was mainly attributed to the evaporation of adsorbed water from the wood samples. In comparison with the untreated wood, the treated wood displayed a similar pattern of TGA curve up 220 °C, but the lower rate of weight loss was observed and eventually higher amount of residue remained. As indicates in Fig. 26, samples PVA1/1C/10M has the lowest percentage decomposition of weight in the first stage of decomposition. The additives of PVA and nanoclay to the wood component increase the thermal stability compared to the untreated wood. For the modified wood, it was verified that the maximum degradation rate was shifted to a higher temperature compare to the UT. Means that, the presence of the PVA and nanoclay improved the thermal stability and increased the degradation temperature of the wood sample. The increase in thermal stability in wood component loaded with nanoclay may be attributed to the presence of silicate layers which acted as a barrier and delayed the diffusion of decomposed volatile products throughout the composite (Rahman et al. 2013). According to Wang and Chai (2012), the distinct weight losses were due to oxidation and pyrolysis of wood components, in which the hemicellulose is most thermally unstable, follow in turn by cellulose and lignin.

Thus, the second stage is between 280 and 400 °C with the total content loss about 67% in the dry ash free. Also, it is initial decomposition of wood component and directly related to the formation of volatile substances from hemicellulose, cellulose and lignin decomposition (Viet and Tho 2017). It can be observed from Fig. 27, the curve of treated woods was shifted to the right, indicating the slow rate of decomposition on the wood component. This was due to the presence of the nanoclay interact

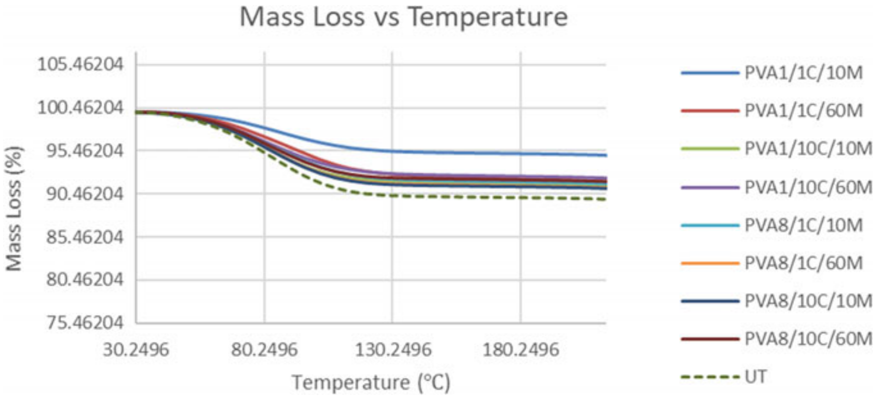


Fig. 26 First stage of decomposition of untreated wood and treated wood

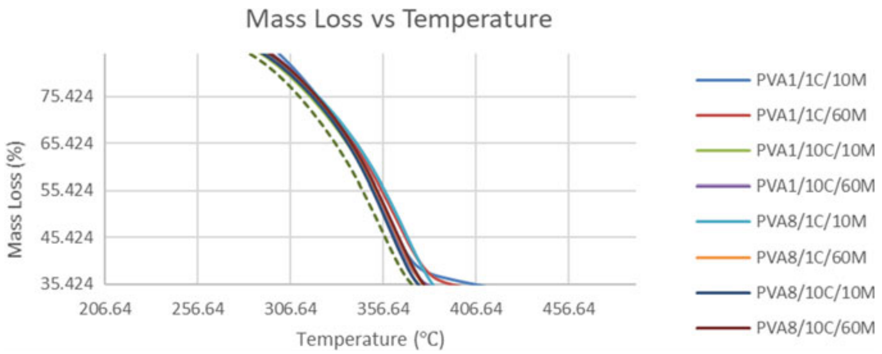


Fig. 27 Second stage of decomposition of untreated wood and treated wood

with the wood cell wall by decreasing the rate of decomposition of the wood. In the last stage, known as continuous decomposition of lignin at a higher temperature to 600 °C and it almost does not have dramatically change of percentage decomposition. The weight loss curves of various samples at different temperatures are simplified in Table 6. In all samples, a slow decrease in weight loss below 70 °C was observed which was due to the removal of some moisture contained in the samples. Also, all samples decomposed dramatically in the temperature range of 280–400 °C. Similar results of acacia wood degradation at temperatures range presented in the pieces of literature (Viet and Tho 2017). The TGA curve of UT showed a single-mass loss step with maximum degradation rate occurred around the temperature of 300–360 °C.

Table 6 shows the decomposition temperature at different weight loss and residual weight (RW%) for untreated wood and treated wood. It was observed that the increases decomposition temperature value will reduce the mass of the wood component. From Table 6, the initial weight loss of the samples below 100 °C was the highest for sample UT compare to the samples of PVA1/1C/10M, PVA1/1C/60M,

Table 6 Residual mass percentage at different temperature

Samples	Residual weight at different temperature (RW%)					
	100 °C	200 °C	300 °C	400 °C	500 °C	600 °C
UT	92.0563	89.9871	80.1212	31.6058	26.0154	22.0503
PVA1/1C/10M	96.5784	95.0686	84.7805	35.8575	29.9329	27.2826
PVA1/1C/60M	94.7124	92.0458	83.1265	34.7963	28.0998	25.1677
PVA1/10C/10M	93.2781	92.5143	81.9769	31.2696	25.0181	22.4027
PVA1/10C/60M	94.2665	92.4273	83.5413	32.9283	26.8130	24.2782
PVA8/1C/10M	93.8331	91.7744	83.0987	31.3992	25.598	22.5501
PVA8/1C/60M	93.1522	91.3054	82.3998	31.5809	25.5084	22.8314
PVA8/10C/10M	93.0911	91.2207	82.4025	31.2788	25.2457	22.6978
PVA8/10C/60M	93.7695	92.0252	83.3547	31.0803	24.9620	22.4544

PVA1/10C/10M, PVA1/10C/60M, PVA8/1C/10M, PVA8/1C/60M, PVA8/10C/10M and PVA8/10C/60M which 7.94% losses of weight. This is due to PVA forms covalent bond with –OH groups of wood cell and surface modified with nanoclay fills the cavity of the wood cell wall for the treated samples.

Moreover, the percentage residual weight of PVA1/1C/10M treated wood sample shows the highest among the other sample followed by sample PVA1/1C/60M. Means that, the amount of PVA and nanoclay was well distributed on the wood component with the optimum impregnation time which could prevent the passage of volatile decomposed product throughout the wood cell. While the highest degradation among the cases is the untreated sample. This is due to the presence of hydroxyl group and cell cavities in the untreated wood component, which they create a path for moisture movement in the component compare to the treated wood sample the hydroxyl group have been reacted with the addition of PVA and nanoclay that change the properties of the wood. Obviously, arising the decomposition temperature values of treated wood, due to higher loading of PVA and nanoclay. Similar observations were prepared by Yadav and Yusoh (2016), while exploring the effect of organoclay in wood plastic composites. So, by incorporation of PVA and nanoclay in cell wall, showed the cell wall components were likely to be shielded by the fire-retardant, which may hamper the cell wall component from being accessible to oxygen and retard their combustion, thus enhancing the stability of wood.

4.5 Differential Scanning Calorimetry (DSC)

Differential Scanning Calorimetry (DSC) is a technique used to study the energy of the thermal transitions released or absorbed via chemical reactions of the raw wood and treated wood constituents during heating which lead to the behavior of exothermic and endothermic conditions (Rahman et al. 2017). Thermal transitions are the changes that take place in a polymer such as the melting of the crystalline and the glass transition of the substance. Basically, exothermic reaction provides information on sample melting, phase transitions, evaporation, dehydration and pyrolysis. Figure 28 shows the DSC curves of sample UT and treated wood made with different PVA and nanoclay contents.

From the graph, the general idea was obtained about the melting temperature and the interaction between PVA and nanoclay with the wood component. In this study, can lead to information about the mechanical properties of the impregnated wood. It was seen that the composites exhibit three patterns of heat flow. First, a strong endothermic peak observed around 87–102 °C for raw wood and treated wood samples. This occurred corresponds to the evaporation of absorbed water by the raw wood and wood composites. Thus, the water loss peak of treated wood samples is shifted to a higher temperature than raw wood. This tendency of releasing water at higher temperature in the treated wood with different amount of nanoclay could due to the improved of wetting property during impregnation of the wood. However, there was some peak of water loss for treated wood samples were shifted

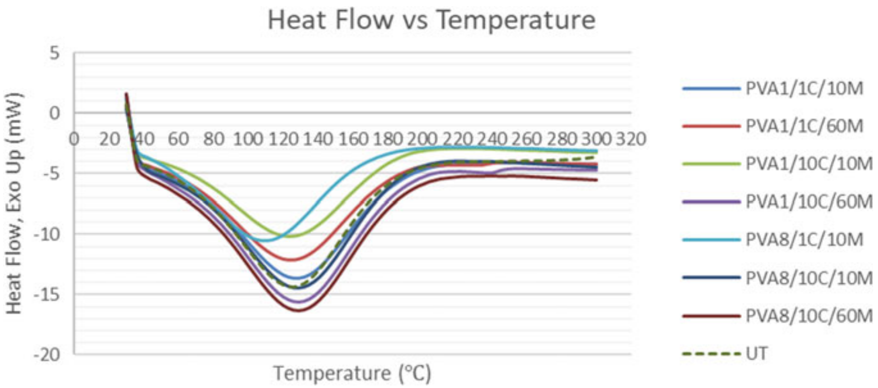


Fig. 28 DSC curves of sample UT and treated wood made with different PVA and nanoclay contents in temperature range of 0–300 °C

at a lower temperature compared to the raw wood. The result may occur due to poor in interfacial interaction of wood cell wall with filler polymer and nanoclay. Moreover, the degradation enthalpy of sample PVA8/10C/60M was very higher than the other composites. Figure 29 indicates the DSC curves of sample UT and treated wood made with different PVA and nanoclay contents in a temperature range of 260–600 °C.

Furthermore, the glass transition of raw wood and treated wood samples cannot be easily detected due to the interference of water evaporation in wood and the presence of unreacted PVA (Rahman et al. 2017). But according to Agronômicas et al. (2010), the glass transition temperature of the wood is the same as the lignin,

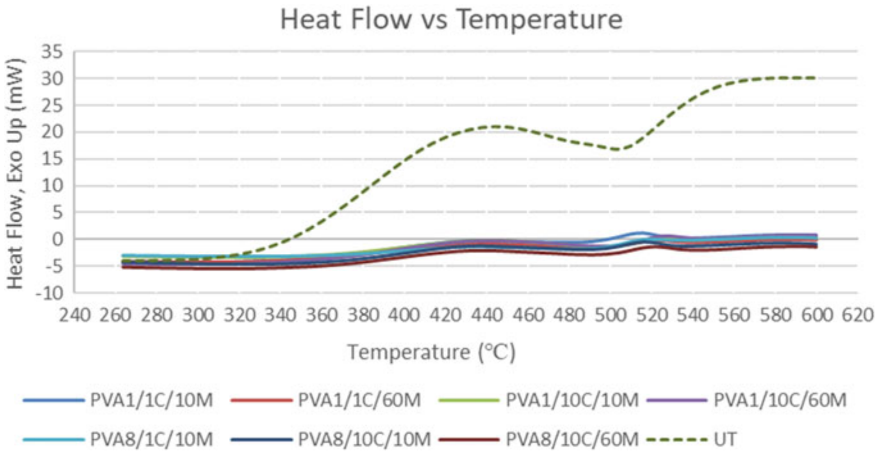


Fig. 29 DSC curves of sample UT and treated wood made with different PVA and nanoclay contents in temperature range of 260–600 °C

which found in between 60 and 200 °C. The next pattern from Fig. 28, showed a very low endothermic peak around 240 and 260 °C which indicates the degradation of cellulose. From the table shows, all treated wood samples had a higher melting temperature (T_m), compared to the raw wood. UT sample had the lowest T_m and sample of PVA8/1C/10M had the highest value of T_m which are 247.91 and 260.41 °C respectively. Figure 29 shows the exothermic peak of the raw samples broad higher than other treated wood samples. According to Rahman et al. (2017), these peaks indicate combustion of lignin and hemicellulose for raw woods and treated wood. This combustion for treated wood was not appreciably occurred due to existing of the polymer and nanoclay which create better interfacial bonding among cell wall of the wood.

4.6 Response Surface Methodology

A mathematical and statistical technique is used to analyse effects of several independent variable by using Response Surface Methodology (RSM) technique (O-1). Basically in numerous cases, the relationship between the response and independent variables are not known. Therefore, RSM was used to determine the optimal treatment variables of impregnating the Acacia wood with Poly(vinyl) alcohol and nanoclay. There are three independent variables which are the concentration of Poly(vinyl) alcohol (PVA), weight of nanoclay (NC) and time of impregnation process (IT). Thus, the response variable was analysed in this study is flexural strength and percentage of residual weight. The RSM was applied using Design Expert Software (State Ease, Design Expert 8). The range and levels each variable was determined by literature reading. The plan of RSM in actual levels of the three independent variables are shown in Table 7.

The design consisted of 8 runs as listed in Table 8. The response functions measured in this study were flexural strength and percentage of residual weight of the sample. Thus, the results of the experimental design were analysed and interpreted using Design Expert Software. The actual result of the desire response in this study was shown in Table 7.

The results of 8 runs to determine the flexural strength and residual weight percentage of the treated wood are tabulated in Table 8. The three factors tested in this study were loading amount of PVA, weight of nanoclay and time for impregnation

Table 7 The range and level of the variables

Factor	Name	Units	Min	Max	Coded low	Coded high	Mean	Std. dev
A	PVA	%	1.0000	8.00	−1 ↔ 1.00	+1 ↔ 8.00	4.50	3.74
B	NC	g	1.0000	10.00	−1 ↔ 1.00	+1 ↔ 10.00	5.50	4.81
C	IT	min	10.00	60.00	−1 ↔ 10.00	+1 ↔ 60.00	35.00	26.73

Table 8 The responses of the parameters used in response surface methodology

Run	Factor A: PVA (%)	Factor B: NC (g)	Factor C: IT (min)	Response 1: flexural strength (MPa)	Response 2: residual weight (%)
1	1	1	60	95.63	25.17
2	1	10	10	118.27	22.40
3	8	10	60	98.14	24.45
4	1	10	60	85.60	24.28
5	8	10	10	115.76	22.70
6	8	1	60	88.08	22.83
7	1	1	10	110.72	27.28
8	8	1	10	115.76	22.50

process of the wood. From Table 8, the highest flexural strength is ranged from 85.56 to 118.27 MPa. While for the 2nd response, the range of residual percentage ranged from 22.40 to 27.28%. The highest value of flexural strength was 118.27 MPa under test condition of 1% PVA loading, 10 g addition of nanoclay and 10 min of impregnation process of the wood. Furthermore, the highest percentage of residual weight was 27.28% under test of 1% of PVA, 1 g of nanoclay and 10 min for wood impregnation process. Meanwhile, the lowest flexural strength value was 85.56 MPa with formulation of 1% of PVA, 10 g of nanoclay and 60 min of impregnation process. Thus, the lowest percentage of residual weight of treated wood from TGA analysis was 22.40% under test condition of 1% PVA loading, 10 g of nanoclay and 10 min for wood impregnation process. According to Rowell (2006), when polymer and nanoclay used as a reinforcing material in wood component, it will improve the mechanical properties and thermal stability of the treated wood.

The collected data was analysed using software Design Expert 11. All the responses were analysed using analysis of variance (ANOVA) and regression analysis for model fitting the significance of the coefficient terms. The analysis of variance (ANOVA) for the design model of flexural strength and percentage of residual weight are presented in Tables 9 and 10 respectively.

The ANOVA demonstrated that the regression model of flexural strength was highly significant as the F-test had a very low probability value. There is only a 1.85% chance that an F-value this large could occur due to noise. The P-values for flexural response is less than 0.0500 indicate model terms are significant. Thus, in this study, Factor C is a significant model term. The p values which greater than 0.1000 indicate the model terms are not significant. It can conclude, the Factor C give higher significant effect on the flexural strength than the other two independent variable. If there are many insignificant model terms, model reduction may improve the model. Therefore, it can be said that time for impregnation process, had the greatest effect on flexural strength. The estimated models built for flexural strength are represented by Eq. (1). It should be noted that the following equations are only valid within range of tested condition that mentioned in Table 7. For flexural strength, the model equation

Table 9 Analysis for variance for the design model of flexural strength

Source	Sum of squares	Degree of freedom	Mean square	F-value	p-value	
Model	1097.93	3	365.98	11.86	0.0185	Significant
A-PVA	7.13	1	7.13	0.2308	0.6560	
B-NC	7.12	1	7.12	0.2307	0.6561	
C-IT	1083.68	1	1083.68	35.10	0.0041	
Residual	123.48	4	30.87			
Correlation total	1221.41	7				

Table 10 Analysis for variance for the design model of residual weight percentage

Source	Sum of squares	Degree of freedom	Mean square	F-value	p-value	
Model	7.90	3	2.63	0.8517	0.5339	Not-significant
A-PVA	5.52	1	5.52	1.79	0.2523	
B-NC	1.95	1	1.95	0.6308	0.4715	
C-IT	0.4267	1	0.4267	0.1380	0.7291	
Residual	12.37	4	3.09			
Cor total	20.26	7				

is as follows:

$$Flexural\ Strength = 117.41655 + 0.269639A + 0.209692B - 0.465549C \quad (1)$$

From Table 9, the model F-value of 0.8517 implies the model is not significant relative to the noise. There is about 53.39% chance that an F-value this larger could occur due to noise. Moreover, there are no significant model terms because p-value are greater than 0.100. Hence, model reduction may improve the model. The estimated linear model built for percentage of residual weight methods is represented by Eq. (2). The following equation is valid within the range of tested condition as mentioned in Table 7.

$$\begin{aligned} Percentage\,of\ Residual\ Weight = 25.30026 - 0.237379A \\ - 0.109711B + 0.009238C \end{aligned} \quad (2)$$

A graphical representation of the model quality is shown in Fig. 30. The predicted value in y-axis versus actual value in x-axis for flexural strength. The fitting of the models was checked by determination coefficient values (R^2). The R^2 value for flexural strength analysis and percentage of residual weight was 0.8989 and 0.3898

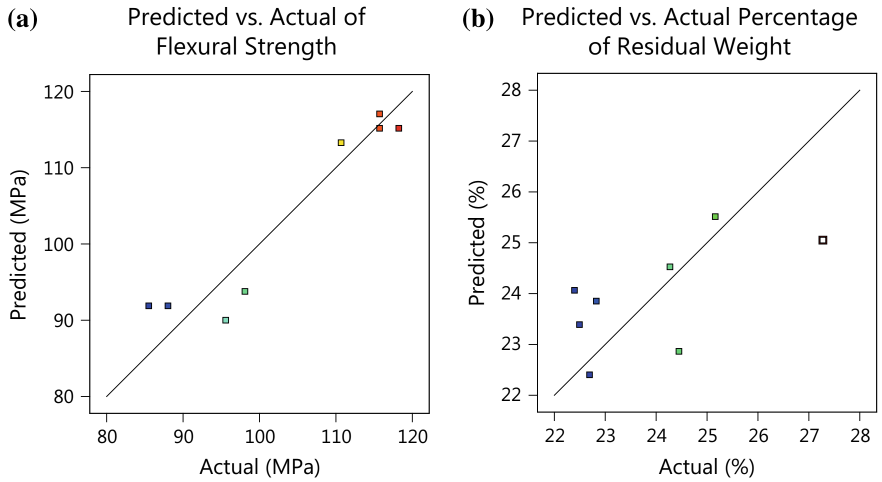


Fig. 30 Correlation of predicted responses versus experimental response: **a** flexural strength and **b** percentage of residual weight

respectively. These R^2 values indicated that 10.11% of the flexural strength variation and 61.02% of the impact strength variation were not explained by the models.

4.6.1 Analysis of Response Surfaces

The 3D response surface plots and contour plots of the combine effects of the independent variables of PVA loading, amount of nanoclay and time for impregnation process on flexural strength and percentage of residual weight are shown in Figs. 31 and 32. In the current study, 3D response surfaces were obtained by keeping one of the variable constants at a zero level while varying the other two variables. Figure 31b and c demonstrate that the time for impregnation process had the most significant effect on the tensile strength, followed by PVA loading and nanoclay loading.

Figure 31b illustrated that flexural strength was slightly decrease with an increase in time for impregnation process at constant PVA loading of 2 wt%. As shown in Table 8, the highest flexural strength was obtained when the impregnation time in the lowest level and the amount of nanoclay at the highest level. The decrease in flexural strength at the longest impregnation time can be due to the degradation of the cellulose. Meanwhile, the value of flexural strength always above 100 MPa even in the minimum and maximum amount of PVA loading and nanoclay as demonstrate in Fig. 31a. Meanwhile, the effects of independent variables on the percentage of residual weight are shown in Fig. 32.

As shown in Fig. 32a, the residual weight of treated wood increase with the decrease amount of PVA loading and nanoclay. However, the highest residual weight showed when the amount nanoclay was 5 g at 2% of PVA concentration. Besides

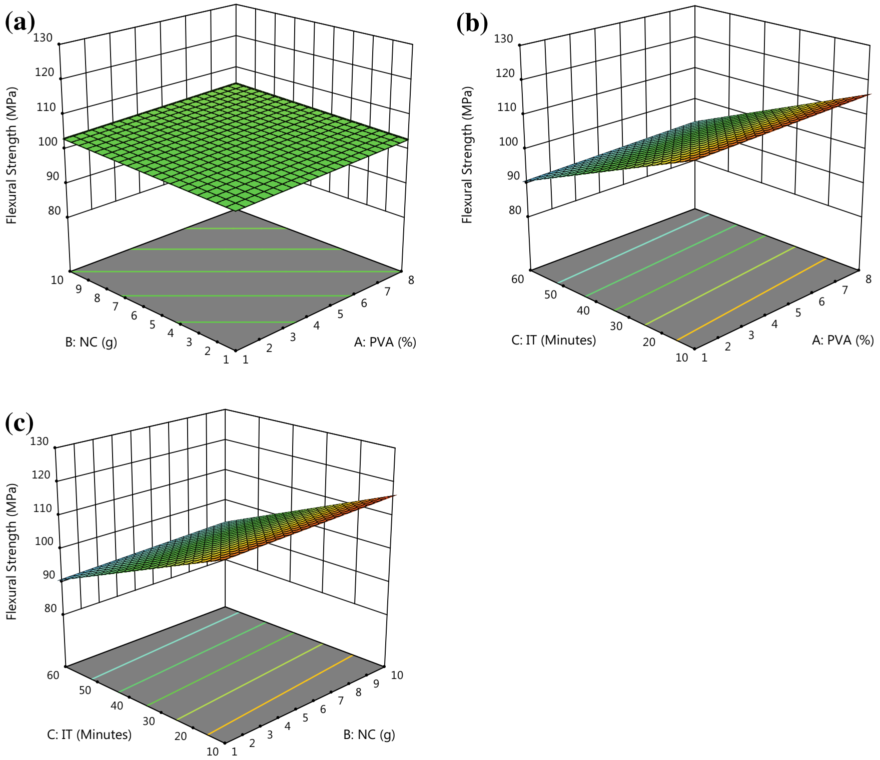


Fig. 31 Response surface plots of the combined effects of the independent variables on flexural strength of treated acacia wood: **a** NC and PVA at fixed value of IT, **b** IT and PVA at fixed value of NC and **c** IT and NC at fixed value of PVA

that, the increase of time impregnation with the lowest concentration of PVA loading, shows the increasing amount of residual weight which resulted from Fig. 32b. A maximum residual weight of 25% was attained at 1% concentration of PVA with 60 min of impregnation time. Figure 32c shows shat the residual weight increase with decreasing amount of nanoclay and increasing the time of impregnation process of the wood. The maximum residual weight was 24.6% with condition test of 1.5 g of nanoclay and 55 min of impregnation time.

4.6.2 Optimization of the Experiments

Response surface methodology (RSM) was used to optimize the conditions for the preparation of wood composites. The design experiment was carried out using Design Expert 11. In the optimization selection, there were three factors for a goal to construct the desirability indices which is concentration of PVA loading, amount of nanoclay and impregnation time. The goals for both flexural strength and residual weight

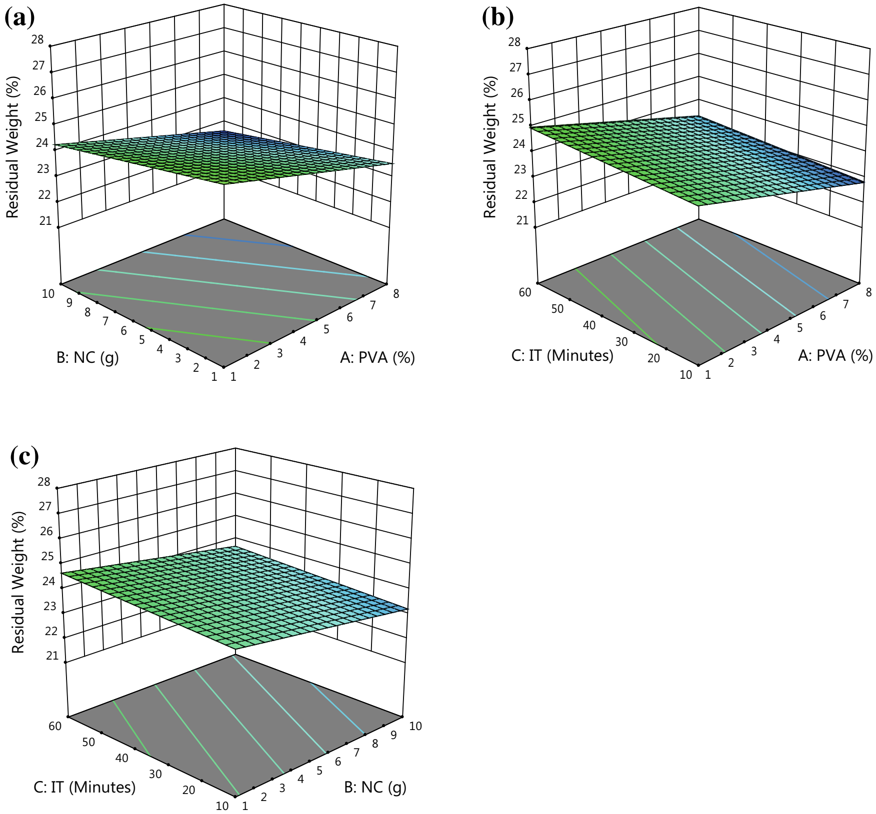


Fig. 32 Response surface plots of the combined effects of the independent variables on residual weight percentage of treated acacia wood in TGA analysis: **a** NC and PVA at fixed value of IT, **b** IT and PVA at fixed value of NC and **c** IT and NC at fixed value of PVA

was to maximize the strength and increase the weight of the wood residue with the minimum amount of independent variables.

Therefore, the target value of the responses was the highest values from the experimental data obtained. The acceptable values of the desirability function were the values close to 100%. So, from the data simulation in Fig. 33, the mechanical properties and thermal stability of the treated wood compromised with 1% of PVA loading, 1 g amount of nanoclay and 10 min of impregnation time with 85.6% desirability. These levels of independent variables yield the highest responses of flexural strength and residual weight which is 113.24 MPa and 25.0456% respectively. To obtain materials with improved mechanical properties, the efficient dispersion of one phase into the other is required.

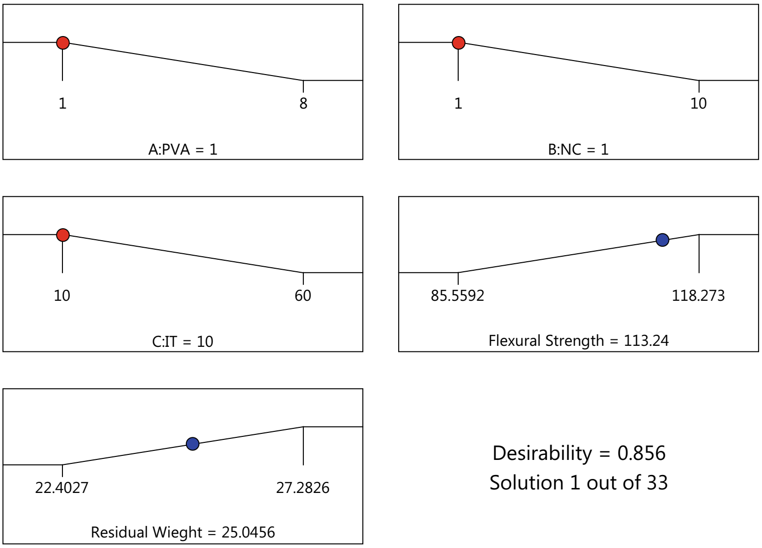


Fig. 33 Optimum condition of the independent variables and the responses of the treated wood

4.7 Summary

The incorporation the PVA and nanoclay with wood cell indicates better improvement in mechanical properties, morphology and thermal stability compare to untreated wood. The properties of the wood sample were tested and the results were obtained and analyzed. The discussion based on the results had been made based on the previous researches. Based, on the discussion and analysis of the results, the improvements and enhancements on the properties of the treated wood had been shown but not all the prepared wood composites showed the massive improvement in their mechanical properties. morphology, chemical structures and thermal stability. The improvement highly depends on the nanofillers loading and also amount of time for impregnation process to take place. The highest sample that posses highest flexural strength is sample PVA1/10C/10C. Also, this sample showed the smoothest surface in the wood component among the other wood composites. In the thermal analysis, sample PVA1/10/10C showed the highest percentage of residual mass, which mean rate of degradation of this sample is lower than the other samples. Thus, PVA and nanoclay is the most promising nanofillers that can be used for reinforcement of Acacia wood. The response surface methodology was successfully applied to determine the optimal operational conditions for high quality of Acacia wood especially in thermal stability and flexural strength improvement. The experimental values were in good agreement with the predicted values and the models highly significant with correlation coefficients between 0.39 and 0.89. It was also found impregnation time had the most significant effect on the responses.

5 Conclusion

Acacia wood that incorporated with PVA and nanoclays have shown enhancement in their properties and are widely used in various fields especially for furniture, construction and interior design. This study investigated the effect of Poly(vinyl) alcohol (PVA) and nanoclay as reinforcing agent on the mechanical properties, morphological and thermal properties of Acacia wood. Also, the impregnation time of the Acacia wood in vacuum chamber also a factor was being observed in this study. The incorporating of impregnation time affect the behavior and result of the impregnated woods. Also, different amount of PVA and nanoclay showed different performance in terms of properties and characteristics. Several tests are done for analyzation purpose which are Three Point Bending Testing, Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC).

Sample PVA1/10C/10M showed a higher value of flexural strength among the other treated wood. The second high of flexural strength value was sample PVA8/1C/10M and PVA8/10C/10M which nearly similar about 115 MPa. However, the longer impregnation time of sample showed the lower value of flexural properties even with same the formulation of chemical preparation for impregnation. Thus, it can be concluded that the minimum time with the desired formulation of chemical indicates high mechanical properties for treated wood. In FTIR study clearly shows the intensity transmittance of the O–H for treated wood is lower than the raw wood. To explain this, it is hypothesized that the polarity of the (–OH) in wood component was reduced by the chemical reaction between the impregnated chemical and a hydroxyl group. Besides that, it was observed that the existence of Silica Oxide (Si–O) from the nanoclay was proven with the peak formation in wavelength between 1000 and 1110 cm^{-1}. This showing the interaction of silicate and the wood cell wall is occurred during the impregnation process. From the discussion on thermal analysis, sample PVA1/10C/10M contain a high amount of residual percentage which is 27% when heated up to 600 °C. The different between untreated wood samples is about 5% residual weight at 600 °C. From this work, it has been concluded that nanoclay are promising materials for the improvement of the mechanical properties and dimensional properties of wood.

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Tensile, Flexural and Impact Strength of Acacia Wood Bio-composites

P. L. Nyuk Khui, Md Rezaur Rahman, S. Hamdan, M. K. B. Bakri, E. Jayamani and A. Kakar

Abstract This chapter explores the tensile, flexural and impact strength of acacia wood biocomposites. The tensile strength of U-AW-PLA and M-AW-PLA bio-composites at different fiber loadings show that 15wt% fiber loading increment 23% compare to pure PLA. Flexural strength for AW-PLA bio-composites was achieved at 15wt% fiber loadings for both U-AW-PLA and M-AW-PLA bio-composites. The percentage of decrement for 15wt% U-AW-PLA is about 1% than the pure PLA. The impact strength for AW-PLA bio-composites was achieved at 15wt% fiber loadings for both U-AW-PLA and M-AW-PLA bio-composites. The percentage of increment for 15wt% U-AW-PHA is about 6% than the pure PHA whereas 15wt% M-AW-PHA is about 4% than the 15wt% U-AW-PHA. The polymer blends (PLA/PHA) bio-composites created desirable mechanical properties (tensile, flexural and impact strength) than the PLA and PHA polymer bio-composites itself due to the interlocked structure.

Keywords Tensile strength · Flexural strength · Impact strength · Mechanical properties

1 Introduction

Acacia's are classified as a hardwood. *Acacia mangium* (AM) or *Acacia auriculiformis* (AA) is a Leguminous tree species of the sub-family *Mimosoideae*. These species are natively and originally found at the North Queensland in Australia, the Western Province of Papua New Guinea and the Moluccas East/Islands of Indonesia

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P. L. Nyuk Khui · Md R. Rahman (✉) · S. Hamdan · M. K. B. Bakri
Faculty of Engineering, Universiti Malaysia Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

E. Jayamani
Faculty of Engineering, Computing and Science, Swinburne University of Technology, Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

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(Doran and Turnbull 1997). In lowland areas, AM is important and usually used as a multipurpose tropical tree. This species was introduced back in 1967 in Sabah as a firebreak barrier. In Sarawak, one million hectares of land were reserved for the forest's replantation and commercialized program before the year of 2020 (PERKASA 2009). It is part of efforts to meet the current and future needs of raw materials demand for the timber industry as well as to preserve and conserve the natural forest.

In the timber industry, these species were chosen due to its fast growth, favorable local climatic conditions, and potential of utilization in a variety of applications. Acacia wood (AW) species are also known to be used to restore soil conditions during its rehabilitation and reforestation (Inagaki and Titin 2009; Yamashita et al. 2008; Yang et al. 2009). AW is good for carpet floors, turning, cabinetry, and furniture products. It is also applicable to be used for plywood, particleboard, fence posts, wood, charcoal and pulp. As for its non-timber products, it can be used for adhesive, ornamental and shade for roads, adhesives, and other uses of urban forestry. The leaves can be used as fodder for livestock and fertilizer. AW grain texture is generally fine, uniform and do not have a distinctive odor or taste, most of it is interlocked with a medium luster.

However, AM plantations are known to face a few setbacks. They are susceptible to heart rot damage (Hashim et al. 1990) and are unsuitable for sawing and peeling (Weinland and Zuhaidi 1990). To overcome these setbacks, considerable efforts were done by planting new hybrid and clones of acacia. In Sarawak, two new breeds have been introduced, which include, mixed species of AM and AA, and 2nd generation AM. The mixed species of AM and AA are also known as Acacia Hybrid (AH). They have more desirable properties than their previous generations, which include lesser tapering, faster growth, straighter bole, and higher resistance to heart rot. AH's characteristics are maintained through its mass propagated cuttings. The 2nd generation AM also known as AM Super Bulk is improved due to the agricultural cultivation program, which propagated by seeds and has a superior performance in terms of resistance to heart rot, stem form, growth rate, and high volume. The large number of woods from previous generation AM is now being neglected by the market, due to its inferior physical and mechanical properties. Thus, this contributes towards a significant amount of waste. More research regarding the handling of waste are required, to prevent this problem from occurring.

Most of natural fiber contains mainly cellulose, hemicelluloses and lignin structure (Li et al. 2007). Anhydro-D-glucose is the elementary unit of cellulose macromolecule. It contained three types of hydroxyl (–OH) group (Khan and Ahmad 1996). Thus, made the fibers hydrophilic in nature causing their moisture content to increase, which cause faster degradation (Bledzki et al. 1996). Hydrophilic nature of the natural fibers causes undesirable adhesion, which loses it strength (Herrera-Franco and Valadez-Gonzalez 2005). According to Seki (2009), pectin and waxy substances hinder the –OH group from reacting with polar matrices and forming mechanical interlocking adhesion with non-polar matrices. Thus, chemical modifications are used to remove the non-cellulosic constituent to obtain reactive –OH groups for interacting with the polymer (Li et al. 2007; Mohanty et al. 2004). Many chemical modification techniques can be applied on the fibers, such as alkaline, esterification, benzoylation, benzene diazonium salt (BDS), silane, permanganate, peroxide, isocyanate, etc. (Abdelmouleh et al. 2007; Aggarwal et al. 2013; Asumani et al. 2012; Bledzki et al. 1996; Corrales et al. 2007; Freire et al. 2008; Goripathi et al. 2012; Haque et al. 2009; Hong et al. 2008; Joseph et al. 1996; Kabir et al. 2010; Misra et al. 2002; Nair et al. 2001; Paul et al. 1997; Rong et al. 2001; Singhal and Tiwari 2014; Wang et al. 2009; Zaman et al. 2010).

Currently, most of the polymer industries produced mass production of non-environmentally friendly type of polymers, which is based on petroleum sources, such as low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl chloride (PVC) (Siddiqui et al. 2008). Most of these materials are non-biodegradable, non-recyclable, non-renewable and non-reusable, which are one of the major problems of materials disposal that contributed to environmental pollution (Siddiqui et al. 2008). To overcome this problem, environmentally friendly types of polymers were introduced. Few examples of biodegradable polymer or bio-polymer are; poly lactic acid (PLA), polyhydroxyalkanaotes (PHA), and poly glycolic acid (PGA) (Averous and Pollet 2012).

Clay is widely used as nano-enhancement due to its abundant resource, environmentally friendly, low cost, understandable intercalation chemistry, high surface area and high surface of reactivity (Deka and Maji 2010; Dewan et al. 2013; Tjong 2006). Major advantage of using clay as a nano-enhancement in polymer to create bio-composites was due to its unique dispersion, when small amount of it were added in the polymer (usually less than 10 wt%), improved the mechanical properties of the bio-composites (Hetzer and Kee 2008). In industry point of view, the used of clay reduced the amount of cost needed to improve the bio-composites properties compared using an enhancement agent (i.e. coupling agent) which is usually costly (Nourbakhsh and Ashori 2009).

2 Experimental

2.1 Materials

The Acacia wood (AW) from *Acacia mangium* species was provided by Sarawak Forestry Corporation Sdn. Bhd. (SFCsb), Kuching, Sarawak, Malaysia. The AW fibers lengths are less than 3.5 mm. It has a density range of 420–483 kg/m^3 (in green soaked volume) and 500–600 kg/m^3 (in dry condition) (Logan and Balodis 1982; Peh et al. 1982; Peh and Khoo 1984; Razali and Kuo 1983; Wang et al. 1989). Naturally grown AW has a specific gravity (SG) of 0.56 kg/m^3, while the re-plantation grown AW has a lower SG of 0.40–0.45 kg/m^3 (National Research Council 1983). However, in Borneo, the SG can specifically be obtained around 0.43–0.47 kg/m^3 for 6-years old AW (Sining 1989).

For naturalization modification technique, two types of chemicals were used; acid and alkaline. The pure acetic acid (CH_3COOH) solution was obtained from Fisher Scientific (UK), Ltd., known by the product name "Acetic Acid, ACROS Organics™". Powdered sodium bicarbonate ($NaHCO_3$) was obtained from Fisher Scientific (UK), Ltd., known by the product name "Sodium Bicarbonate, ACROS Organics™". Polyhydroxyalkanoates (PHA) was obtained from Jinan Pu Dong Import and Export Trading (China) Co. Ltd. with chemical abstracts service (CAS) number "718630-59-2". It had a density of 1.2–1.4 kg/m^3 and contained 90% bio-based carbon according to ASTM D6866-16 (2016) standard. Pure grade powdered PLA was obtained from Shenzhen Esun Industrial (China) Co. Ltd. with product code "Al-1001". It had 100% bio-based carbon according to the ASTM D6400-12 (2012) standards. It had a density of 1.25 kg/m^3 and melt flow index (MFI) of 10–12 g/10 min.

2.2 Fiber Preparation

For unmodified fiber, the AW fibers were washed and rinsed with distilled water. Later, it was dried in an open ventilated oven at 80 °C for 24 h. The ventilated oven model "ECOCELL EC55" equipped by MMM Medcenter Einrichtungen (Germany) GmbH was used in this process.

This process involved the use of two different man-made bio-based chemicals. The AW fibers were soaked for 24 h inside 1.5 L of 5 wt% of acetic acid solution (CH_3COOH) mixed with distilled water. After that, 90 g of sodium bicarbonate (NaHCO3) was poured into the soaked AW for 24 h. The reactions of all possible chemicals are as below:

$$Na_2CO_3 + 2CH_3COOH \rightarrow 2CH_3COONa + H_2O + CO_2 \tag{1}$$

$$Na_2HCO_3 + CH_3COOH \rightarrow NaCH_3COO + H_2O + CO_2 \tag{2}$$

$$Wood{-}OH + CH_3COONa \rightarrow Wood{-}O{-}NaH + CH_3COO \tag{3}$$

The short AW fibers were filtered and rinsed using distilled water to neutralized remaining chemicals attached to the AW fibers. Lastly, it was dried in an open ventilated oven at 80 °C for 24 h. All the materials obtained were concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.3 Fabrication of the Bio-composites

Pure PLA, PHA, and PLA/PHA (1:1 ratio of PLA/PHA) with addition of different fiber loadings (i.e. 5, 10, 15 and 20 wt%) are fabricated. The mixtures of the ingredients were done using a drying mixer machine. It was mix blend for about 5 min. The molds used for the bio-composites fabrication, were designed according to ASTM D638-14 (2014) for tensile strength, ASTM D790-17 (2017) for flexural strength, and ASTM D4812-11 (2011) for impact strength. The molds were cleaned and waxed before they were filled in with the mixture of composite materials (i.e. fiber, polymer and nano-enhancement). Waxes were applied on the molds. The releasing wax contained carnauba that eases the removal of samples from the mold. Aluminum foils were placed in between the plates, which made the samples easier to be removed from the molds and reduced the formation of bubbles in the samples. The mixtures of composite materials were placed in the mold frame according to the fiber ratio. The mold was compressed using a hot press hydraulic machine model 'LS-22071' equipped by Lotus Scientific (Malaysia) Sdn. Bhd. under pressure of 1000 psi (6.89 MPa). It was then placed in the hot press machine and heated at 150 °C for 6 min. The samples were then quenched in water at room temperature, according to ASTM E41-92 (2010). The samples were then dried using a hair dryer and were kept concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.4 Methods

2.4.1 Tensile Test

Tensile tests were done according to ASTM D638-14 (2014) standard using Universal Testing Machine instrument, T-Machine Technology (Taiwan) Co. Ltd. model 'UTM107'. The crosshead speed used is 5 mm/min. The tensile strengths were calculated by dividing the maximum load by the cross-sectional area of the sample as below:

$$\sigma_T = \frac{F}{A_o} \tag{4}$$

where, σ_T is the tensile strength at break, F is the load, and A_o is the cross-sectional area. The results of the five samples were chosen and tested for each weight percentage. The results of the five tests were averaged.

2.4.2 Flexural Test

Flexural tests were done according to ASTM D790-17 (2017) standard using Universal Testing Machine instrument. The machine was supplied by T-Machine Technology (Taiwan) Co. Ltd. model 'UTM107'. The crosshead speed used is 5 mm/min. The flexural strengths were calculated for each sample under a load, on a three-point bending setup, based on the following equation:

$$\sigma_F = \frac{3PL}{2bd^2} \tag{5}$$

where, σ_F is the flexural strength at break, P is the load, b is the width, and d is the depth (thickness). The results of the five samples were chosen and tested for each weight percentage. The results of the five tests were averaged.

2.4.3 Impact Test

Impact tests were done according to ASTM D4812-11 (2011) standard using Charpy Impact Tester instrument. The machine was supplied by Lotus Scientific (Malaysia) Sdn. Bhd. model 'LS-22 006-50J'. The angle of release is about a 120° from the sample. The results were recorded after the impact and the energy absorbed by each sample was calculated. The impact strength for 50 J energy absorbed onto the sample is calculated as below:

$$\sigma_I = \frac{E}{A} \tag{6}$$

where, σ_I is the impact strength at break, E is the energy absorbed by the broken sample, and A is the cross-sectional area. The results of the five samples were chosen and tested for each weight percentage. The results of the five tests were averaged.

3 Results and Discussion

3.1 Effects of Fiber Loading on PLA and PHA Bio-composites Mechanical Properties

Figures 1, 2, 3, 4, 5, 6, 7, 8, 9, 10 and 11 shows PLA and PHA polymer with addition of unmodified AW fiber and modified AW fiber as reinforcement in bio-composites. Figure 1 shows the tensile strength of U-AW-PLA and M-AW-PLA bio-composites at different fiber loadings. Based on Fig. 1, the desirable tensile strength for AW-PLA bio-composites was achieved at 15 wt% fiber loading for both U-AW-PLA and M-AW-PLA bio-composites. The percentage of increment for 15 wt% U-AW-PLA is about 23% than the pure PLA. The percentage of increment for 15 wt% M-AW-PLA is about 7% than the 15 wt% U-AW-PLA. Figure 2 shows the stress-strain curves of U-AW-PLA and M-AW-PLA bio-composites at 15 wt% fiber loadings.

Figure 3 shows the tensile strength of U-AW-PHA and M-AW-PHA bio-composites at different fiber loadings. Based on Fig. 3, the desirable tensile strength for AW-PHA bio-composites was achieved at 15 wt% fiber loadings for both U-AW-PHA and M-AW-PHA bio-composites. The percentage of increment for 15 wt% U-AW-PHA is about 30% than the pure PHA. The percentage of increment for 15 wt%

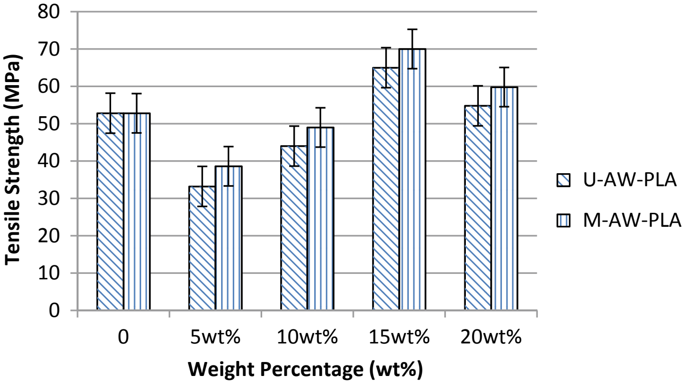

Fig. 1 Tensile strength of U-AW-PLA and M-AW-PLA at 15 wt% fiber loadings

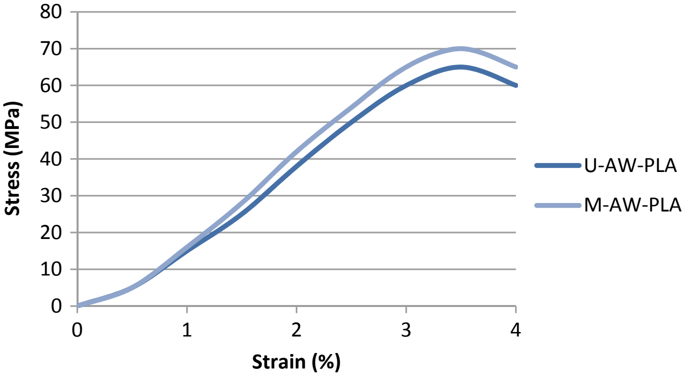

Fig. 2 Stress-strain curves of U-AW-PLA and M-AW-PLA at 15 wt% fiber loadings

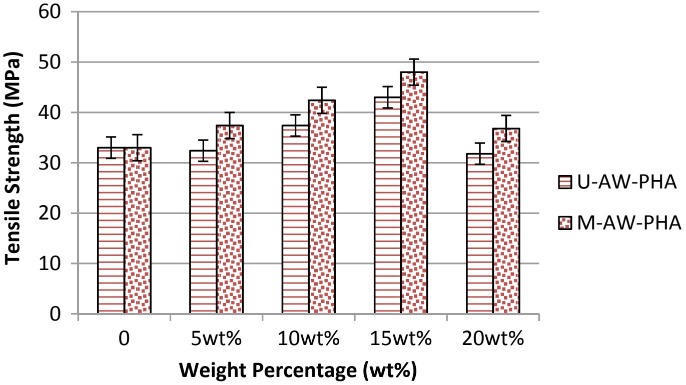

Fig. 3 Tensile strength of U-AW-PHA and M-AW-PHA at different fiber loadings

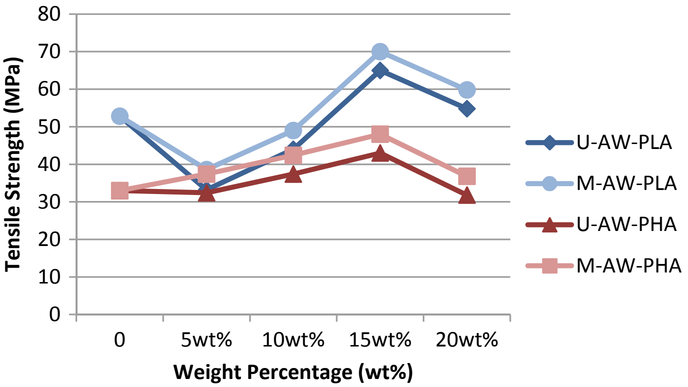

Fig. 4 Comparison of tensile strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings

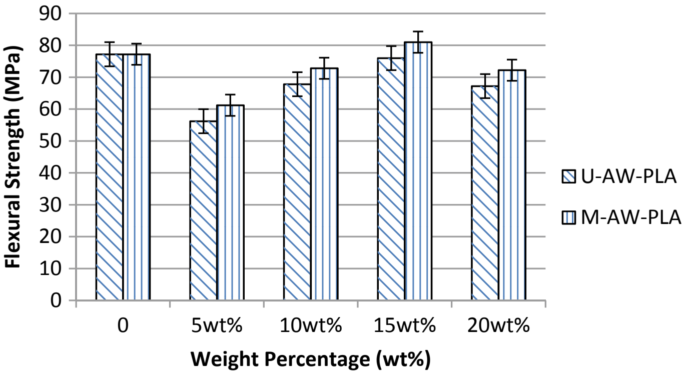

Fig. 5 Flexural strength of U-AW-PLA and M-AW-PLA at different fiber loadings

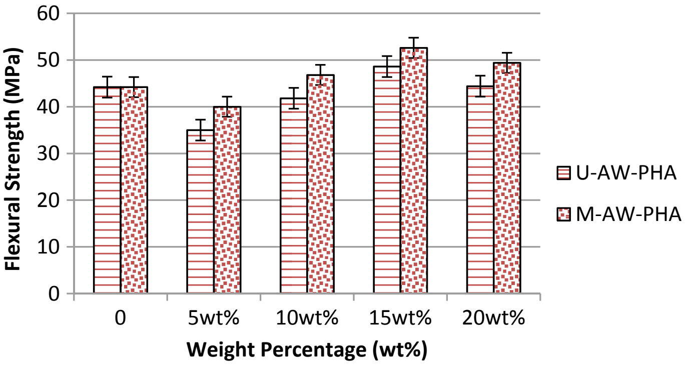

Fig. 6 Flexural strength of U-AW-PHA and M-AW-PHA at different fiber loadings

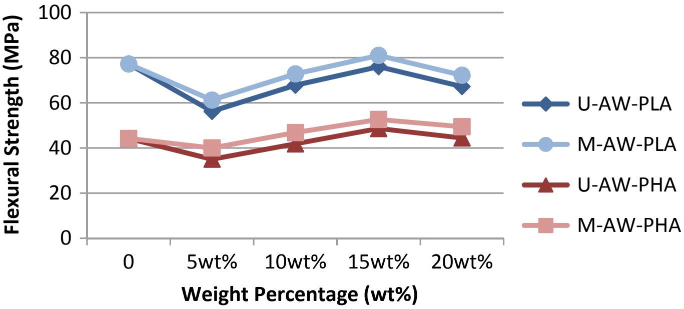

Fig. 7 Comparison of flexural strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings

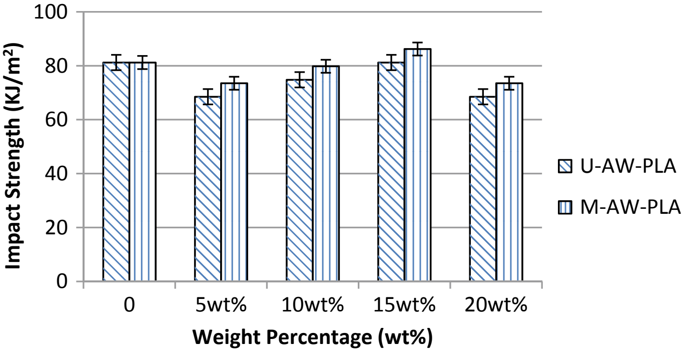

Fig. 8 Impact strength of U-AW-PLA and M-AW-PLA at different fiber loadings

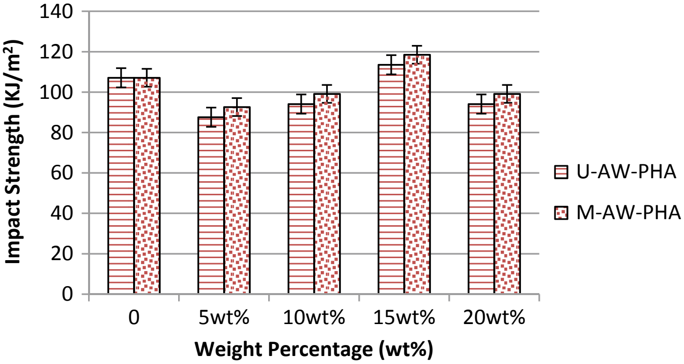

Fig. 9 Impact strength of U-AW-PHA and M-AW-PHA at different fiber loadings

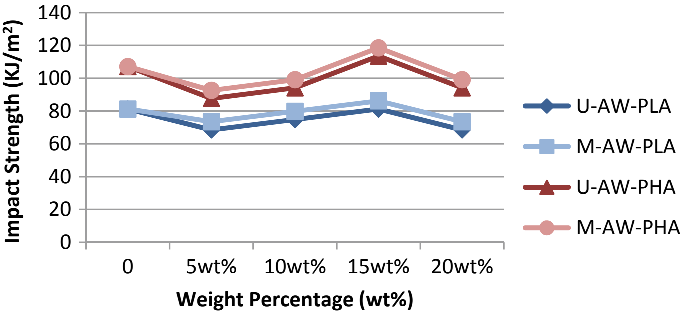

Fig. 10 Comparison of impact strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings

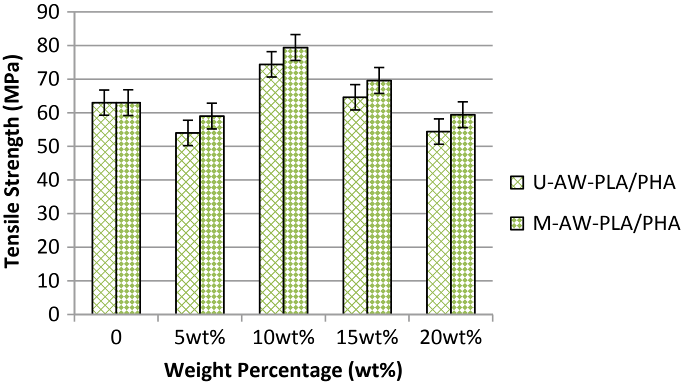

Fig. 11 Tensile strength of U-AW-PLA/PHA and M-AW-PLA/PHA at different fiber weight percentage

M-AW-PHA is about 11% than the 15 wt% U-AW-PHA. Figure 4 shows the comparison tensile strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings.

Figure 6 shows the flexural strength of U-AW-PLA and M-AW-PLA bio-composites at different fiber loadings. Based on Fig. 5, the desirable flexural strength for AW-PLA bio-composites was achieved at 15 wt% fiber loadings for both U-AW-PLA and M-AW-PLA bio-composites. The percentage of decrement for 15 wt% U-AW-PLA is about 1% than the pure PLA. The percentage of increment for 15 wt% M-AW-PLA is about 6% than the 15 wt% U-AW-PLA. Figure 6 shows the flexural strength of U-AW-PHA and M-AW-PHA bio-composites at different fiber loadings. Based on Fig. 6, the desirable flexural strength for AW-PHA bio-composites was achieved at 15 wt% fibre loadings for both U-AW-PHA and M-AW-PHA bio-composites. The percentage of increment for 15 wt% U-AW-PHA is about 9% than the pure PHA. The percentage of increment for 15 wt% M-AW-PHA is about 8%

than the 15 wt% U-AW-PHA. Figure 7 shows the comparison flexural strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings.

Figure 8 shows the impact strength of U-AW-PLA and M-AW-PLA bio-composites at different fiber loadings. Based on Fig. 8, the desirable impact strength for AW-PLA bio-composites was achieved at 15 wt% fiber loadings for both U-AW-PLA and M-AW-PLA bio-composites. The percentage of increment for 15 wt% U-AW-PLA is about 1% than the pure PLA. The percentage of increment for 15 wt% M-AW-PLA is about 6% than the 15 wt% U-AW-PLA. Figure 9 shows the impact strength of U-AW-PHA and M-AW-PHA bio-composites at different fiber loadings. Based on Fig. 9, the desirable impact strength for AW-PHA bio-composites was achieved at 15 wt% fiber loadings for both U-AW-PHA and M-AW-PHA bio-composites. The percentage of increment for 15 wt% U-AW-PHA is about 6% than the pure PHA. The percentage of increment for 15 wt% M-AW-PHA is about 4% than the 15 wt% U-AW-PHA. Figure 10 shows the comparison impact strength for U-AW-PLA, M-AW-PLA, U-AW-PHA and M-AW-PHA at different fiber loadings.

The addition of fiber in the PLA and PHA polymer caused a reduction in the use of expensive polymer, such as PLA and PHA, which make them less costly, especially in the fabrication part of the bio-composites. The increase in the mechanical strength (tensile, flexural and impact strength) of the PLA and PHA polymer bio-composites was due to the increase in the fiber loadings of the bio-composites. The increased amount of fiber in the bio-composites distributed the load and stress uniformly within the bio-composites (Liu et al. 2009). It was noticed that the bio-composites had lower mechanical strength at 5 and 10 wt% due to less amount of fiber incorporated and distributed, which caused an uneven distribution of load and stress within the bio-composites. The undispersed fiber was due to less fiber content, which causes highly localized stress, load or strain to occur in the polymer (Mohanty et al. 2000). While at 20 wt%, the bio-composites showed a decrease in the mechanical strength due to over-distribution of fiber within the bio-composites. This was due to the less polymer use in the bio-composites which act as a binder that attached the fibers together. Less incorporated and over-excessive fiber content cause non-uniform transfer of load and stress within the bio-composites.

Liu et al. (2009) stated that one of the failures in strength was also due to the fiber agglomeration within the polymer bio-composites. While, Mohanty et al. (2000) stated that the desirable fiber content varies with the fiber and polymer ratio, and fiber and polymer interaction. Meanwhile, according to Miah et al. (2011), the effect of the fiber loading is explained by the homogeneity of the fiber and wettability of the polymer. Another factor that may have affected the tensile strength of the bio-composites is the types and characteristics of polymer used. Comparing PLA and PHA, it was observed that PLA had higher tensile strength compared with PHA. This was due to the properties of PLA and PHA itself, which is brittle and ductile, respectively. According to Jiang and Zhang (2017), PLA has single and double bond atoms in it structure, while PHA has a chemical structure that has less carbon atoms compared with PLA, in which reacted with its polyesters. Thus, making PLA a bit more amorphous and brittle compare with PHA, which is a bit more ductile (Wagner et al. 2014).

3.2 *Effect of Polymer Blend (PLA/PHA) on Bio-composites Material Properties*

Figures 11, 12, 13 and 14 shows polymer blend (PLA/PHA) with addition of unmodified AW fiber and modified AW fiber as reinforcement in bio-composites. Figure 11 shows the tensile strength of U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites at different fiber loadings. Based on Fig. 11, the desirable tensile strength for AW-PLA/PHA bio-composites was achieved at 10 wt% fiber loadings for both U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites. The percentage of increment for 10 wt% U-AW-PLA/PHA is about 18% than the pure PLA/PHA. The percentage of increment for 10 wt% M-AW-PLA/PHA is about 6% than the 10 wt% U-AW-PLA/PHA. Figure 12 shows the stress-strain curves of U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites at 10 wt% fiber loadings.

Figure 13 shows the flexural strength of U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites at different fiber loadings. Based on Fig. 13, the desirable flexural strength for AW-PLA/PHA bio-composites was achieved at 10 wt% fiber loadings for both U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites. The percentage of increment for 10 wt% U-AW-PLA/PHA is about 1% than the pure PLA/PHA. The percentage of increment for 10 wt% M-AW-PLA/PHA is about 6% than the 10 wt% U-AW-PLA/PHA.

Figure 14 shows the impact strength of U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites at different fiber loadings. Based on Fig. 14, the desirable impact strength for AW-PLA/PHA bio-composites was achieved at 10 wt% fiber loadings for both U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites. The percentage of increment for 10 wt% U-AW-PLA/PHA is about 22% than the pure PLA/PHA. The percentage of increment for 10 wt% M-AW-PLA/PHA is about 10% than the 10 wt% U-AW-PLA/PHA.

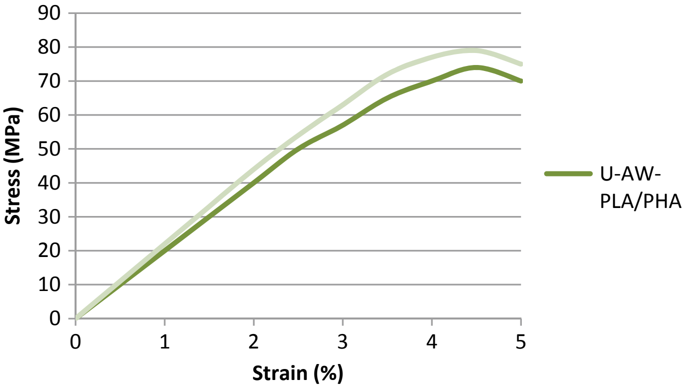

Fig. 12 Stress-strain curves of U-AW-PLA/PHA and M-AW-PLA/PHA at 10 wt% fiber loadings

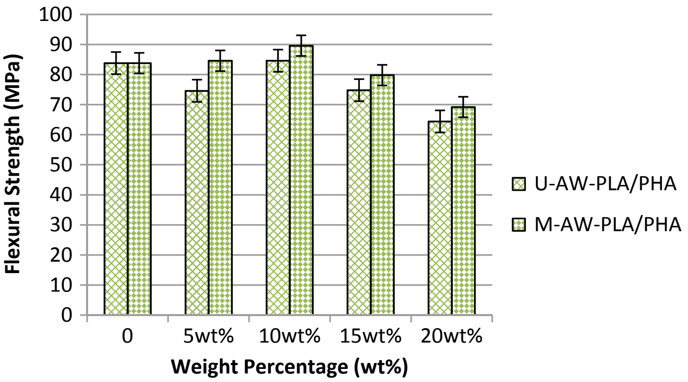

Fig. 13 Flexural strength of U-AW-PLA/PHA and M-AW-PLA/PHA at different fiber weight percentage

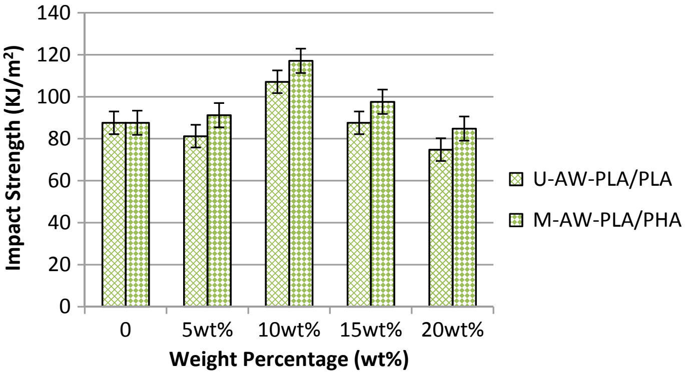

Fig. 14 Impact strength of U-AW-PLA/PHA and M-AW-PLA/PHA at different fiber loadings

The polymer blends (PLA/PHA) bio-composites created desirable mechanical properties (tensile, flexural and impact strength) than the PLA and PHA polymer bio-composites, due to the interlocked structure formed. The extra C=O bond cause it to be stronger than the existing PLA and PHA individually (Jiang and Zhang 2017; Wagner et al. 2014). The stronger desirable tensile strength of polymer blend bio-composites cause reduction on the use of PLA, PHA and fiber itself, which reasoned it to be shifted at 10 wt% fiber loadings when compared with PLA and PHA polymer bio-composites at 15 wt% fiber loadings. Similar results were shown by Loureiro et al. (2015) for their PLA/PHA blends, where proper blend create desirable mechanical strength than its existing PLA and PHA itself, with proper condition being held on (i.e. temperature control, polymer chemical composition, etc.).

4 Conclusion

In this chapter, it is noted that the addition of AW fiber increased the mechanical properties (tensile, flexural and impact strength) of the bio-composites. It was found that the desirable strengths were achieved at 15 wt% fiber loading for PLA and PHA bio-composites and 10 wt% fiber loading for polymer blend (PLA/PHA) bio-composites. Polymer blend create stronger properties than the PLA and PHA bio-composites itself, due to stronger interlockings between PLA and PHA. Chemical modification on the fiber increased the roughness and pores structure of the AW fiber which create stronger interlockings and adhesion between AW fiber and polymer.

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Study of Surface Behavior of Acacia Wood Bio-composites by Morphological Analysis

P. L. Nyuk Khui, Md Rezaur Rahman, S. Hamdan, M. K. B. Bakri, E. Jayamani and A. Kakar

Abstract This chapter describe the surface performance and morphological analysis of acacia wood bio-composites. The SEM images of U-AW-PLA and U-AW-PLA showed a big void between the fiber and polymer. It could be observed that the U-AW has a smooth surface structure whereas the PLA structure was brittle. Impurities influences the properties, where oily and waxy structure of U-AW fiber causes poor adhesion and absorbance between PLA matrix and U-AW fibre. The U-AWPHA and U-AW-PHA shows a void between fiber and polymer and it was observed that the U-AW and PHA structure is smooth and ductile, while U-AW-PHA exhibited the de-bonding between fiber and polymer. Impurities such as the waxy and oily structure of AW fiber influences the properties, hence caused poor adhesion and absorbance between U-AW fiber and PHA. It could be observed from the SEM images that the U-AW and NCHB-PLA/PHA has a smooth and semibrittle/semi-ductile structure. The resulting properties could be caused by impurities such as the oily and waxy structure of U-AW fiber, hence affecting the composite sample by having poor adhesion and absorbance between U-AW fiber and NCHB-PLA/PHA.

Keywords Morphological · Surface · Bio-composites · Wood

1 Introduction

Fiber dispersion is a major factor influencing the properties of composites, the application of short fibers requires good fiber dispersion due to its hydrophilic nature

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P. L. Nyuk Khui · Md R. Rahman (✉) · S. Hamdan · M. K. B. Bakri
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

E. Jayamani
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

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coupling together with hydrophobic matrices (Sanadi et al. 1997). Longer fibers are inclining to cause agglomeration, hence to reduced voids in composites requires better fiber dispersion in the matrix, promoting better interfacial adhesion and bonding (Heidi et al. 2011). Temperature and pressure are some processing parameters in which influences the fiber dispersion. According to Sanadi et al. (1997), the application of steric acid in PP and PE modified the fiber dispersion in the composites, with the addition of MAPP it increases the interfacial adhesion hence influencing more interfacial interaction of the fiber matrix. Similarly grafting fiber modification could also be used to improved fiber dispersion, but it is considered as an expensive option compare to other modifications involving fiber dispersion (Sanadi et al. 1997). Mixing processes such as the single and twin-screw extruder also has the effects of influencing fiber dispersion, it was observed that there were more fiber dispersion occurring in the twin screw extruder as compared to single screw extruder. Although processing causes damaged and reduce the size of the fiber dramatically, it consistently varies according to set temperature and screw configurations (Beckermann and Pickering 2008; Sanadi et al. 1997).

According to numerous studies, composite materials could acquire good mechanical properties if the fiber direction is aligned parallel to the direction of the applied load (Amor et al. 2010; Herrera-Franco and Valadez-Gonzales 2005; Norman and Robertson 2003). Majority of natural fibers are known to have a random alignment as compare to a continuous alignment, where by synthetic fibers are observed to have continuous alignment. Factors such as mold design and matrix viscosity could influence and achieved some form of alignment, especially during injection molding (Joseph et al. 1999). Upon fabrication prior to the matrix impregnation, to achieve more fiber alignment, long natural fibers were manually placed and carded in sheets.

Since 1970s, wrap sinning was a method to align and produce fiber yarns. Through the amount of subsequent processing to form a continuous fiber, short fibers were utilized and converted by strand wrapping around discontinuous fibers (Pickering et al. 2016). Compression molding (CM) could utilized continuous strands from the same types of fiber as the short fibers, and thermoplastics could be used to form the matrix material. The converted thermoplastic fibers could act as a support for the natural fibers and be utilized in the yarn in an aligned direction.

Carpenter et al. (2007) reported that there were improved flexural, stiffness and tensile strength with unidirectional yarn in comparison with conventional twisted yarns for flax epoxy and aligned fiber yarn. According to Khalfallah et al. (2014), continuous fiber tape was produced from an adhesive in the form of pectin extracted from the fibers, by using water mist and dry stretched. To achieve good degree of fiber alignment embedded in the composites similarly as continuous synthetic fibers (i.e. pultrusion, filament, etc.), continuous materials could be utilized. Regarding cellulose chains, cellulose chains have an angle relative to their fiber elemental direction or microfibrillar angle.

Alternative methods such as Dynamic sheet forming (DSF) utilizes the extensive infrastructure of the fiber alignment in the composites, DSF is known to be used as a traditionally method for paper production to align the natural fibers. According to Pickering et al. (2016), short fibers was sprayed through a nozzle suspended in water onto a rotating drum covered with a wire mesh filtering water, hence brings about the fiber alignment via spraying and rotating direction (Pickering et al. 2016). It was report that the improvement in the mechanical properties was comparable to other alternative methods especially with the utilization of short fibers. The mechanical performance and degree of orientation were like synthetic fiber. On the other hand, the natural fiber Young's modulus and strength were observed to be reduce, while the fiber orientation angle increases relative to the testing direction. (Pickering et al. 2016). Brahim and Cheikh (2007) reported that the Alfa fiber reinforced unsaturated polyester (UP) composites exhibited a decrease in strength in relation to the fiber direction (0°) by 69, 29, 22, and 12% at fiber angle of 10°, 30°, 45°, and 90°, and Young's modulus 93, 66, 52 and 41% of similar fiber direction, respectively. According to numerous studies by Baghaei et al. (2014), hemp/PLA composites fiber alignment at 45° and 90° were observed to have 48 and 30% of strength and 53 and 42% of Young's modulus in comparison with the 0° fiber angle.

Morphological study was conducted to investigate and understand the structure of the bio-composites physically, and mechanically. At a microstructure level the morphological study was used to identify the crack, fracture, and etc. on the surface of the bio-composites. (Fornasieri et al. 2011). Examples such as; scanning electron microscope (SEM), light/optical microscope (L/OM), transmission electron microscope, etc. are methods and equipment utilize to investigate and understand the microstructure of bio-composites. Fornasieri et al. (2011) reported some SEM images which showed the interfacial bonding of fiber and matrix and fiber dispersion, where there was poor interfacial bonding due to the pull-out and defects of the fiber in the bio-composites.

Morphological analysis conducted was mostly based upon the fiber rupture, fiber pull out, break-tip, and/or at the fracture of the bio-composites. Reddy et al. (2012) reported that the SEM was utilized to identify the interfacial adhesion of fiber and matrix for coupling agent due to its enhancement, in relations to the improvement of the bio-composites mechanical properties. Verma et al. (2013) verified the concept of utilizing SEM to display the increased spot of fiber in the bio-composites. Perinovic et al. (2010) observe and reported weak fiber and matrix interactions from utilizing a similar method. Alternatively, Ticoalu et al. (2013) has reported that L/OM could be utilized as an alternative method to observe and identify the fiber void/bubble, hollow structure, fiber wall cell, cellulose, hemicellulose, lignin and other impurities.

2 Experimental

2.1 Materials

Acacia wood (AW) from *Acacia mangium* species was provided by Sarawak Forestry Corporation Sdn. Bhd. (SFCsb), Kuching, Sarawak, Malaysia. AW fibre lengths are less than 3.5 mm with a density range of 500–600 kg/m^3 (dry condition) and

420–483 kg/m^3 (green soaked volume) (Logan and Balodis 1982; Peh et al. 1982; Peh and Khoo 1984; Razali and Kuo 1983; Wang et al. 1989). In Borneo, the specific gravity (SG) of AW could be specifically obtained around 0.43–0.47 kg/m^3 for a 6-years old AW (Sining 1989). Naturally grown AW has a SG of 0.56 kg/m^3, while the re-plantation grown AW has a lower SG of 0.40–0.45 kg/m^3 (National Research Council 1983).

Acid and alkaline are the two chemicals used for the naturalization modification technique. Pure acetic acid (CH_3COOH) solution was obtained from Fisher Scientific (UK), Ltd., known by the product name "Acetic Acid, ACROS Organics™". Powdered sodium bicarbonate ($NaHCO_3$) was obtained from Fisher Scientific (UK), Ltd., known by the product name "Sodium Bicarbonate, ACROS Organics™". Polyhydroxyalkanoates (PHA) with a density of 1.2–1.4 kg/m^3 and contained 90% bio-based carbon according to ASTM D6866-16 (2016) standard was obtained from Jinan Pu Dong Import and Export Trading (China) Co. Ltd. with chemical abstracts service (CAS) number "718630-59-2". Pure grade powdered PLA with a density of 1.25 kg/m^3 and melt flow index (MFI) of 10–12 g/10 min, contained 100% bio-based carbon according to the ASTM D6400-12 (2012) standard was obtained from Shenzhen Esun Industrial (China) Co. Ltd. with product code "Al-1001".

2.2 Fiber Preparation

In preparations for the unmodified fiber, AW fiber were washed and rinsed with distilled water. Placed in an open ventilated oven at 80 °C for 24 h for drying. The ventilated oven utilized for drying was a model "ECOCELL EC55" equipped by MMM Medcenter Einrichtungen (Germany) GmbH.

Fiber preparation process further involved the use of two different man-made bio-based chemicals. AW fibers were soaked for 24 h inside 1.5 L of 5 wt% of acetic acid solution (CH_3COOH) mixed with distilled water. After that, 90 g of sodium bicarbonate ($NaHCO_3$) was poured into the soaked AW for 24 h. The reactions of all possible chemicals are as below:

$$Na_2CO_3 + 2CH_3COOH \rightarrow 2CH_3COONa + H_2O + CO_2 \tag{1}$$

$$Na_2HCO_3 + CH_3COOH \rightarrow NaCH_3COO + H_2O + CO_2 \tag{2}$$

$$\text{Wood-OH} + CH_3COONa \rightarrow \text{Wood-O-NaH} + CH_3COO \tag{3}$$

The short AW fibers were filtered and rinsed using distilled water to neutralized residual chemicals attached to the AW fibers. Finally, the short AW fibers was dried in an open ventilated oven at 80 °C for 24 h. All the materials obtained were concealed at room temperature conditioning, as according to ASTM E41-92 (2010).

2.3 Fabrication of the Bio-composites

Bio-composites fabricated were; Pure PLA, PHA, and PLA/PHA (1:1 ratio of PLA/PHA) with addition of different fiber loadings (i.e. 5, 10, 15 and 20 wt%). Mixtures of materials were blended using a drying mixer machine, for about 5 min. Bio-composites were fabricated with molds designed according to ASTM D790-17 (2017) for flexural strength, D638-14 (2014) for tensile strength, and ASTM D4812-11 (2011) for impact strength. Prior to the fabrication, the molds were cleaned and waxed coated before filling the mixture of composite material (i.e. fiber, polymer and nano-enhancement) inside the mold. The wax coating place onto the mold is a releasing wax containing carnauba, in which assist the removal of samples fabricated from the mold. Aluminum foils were placed in between the mold plates which reduces the formation of bubbles and eases the removal of samples. Composite material mixtures were placed into the mold according to the different fiber loadings and the mold was compressed using a hot press hydraulic machine model 'LS-22071' equipped by Lotus Scientific (Malaysia) Sdn. Bhd. under pressure of 1000 psi (6.89 MPa). The compressed mold in the hot press machine was heated at about 150 °C for 6 min, after that the samples were quenched in water at room temperature according to ASTM E41-92 (2010). Finally, samples were dried to remove excess water and kept concealed at room temperature, according to ASTM E41-92 (2010).

2.4 Methods

2.4.1 Scanning Electron Microscopy

Prior to the Scanning Electron Microscopy (SEM), samples were prepared by slicing into small pieces and mounted on aluminum stubs. Samples were coated finely with metal particles (i.e. gold) for about 1–2 min, depending on the size of the samples prepared. The coating process was conducted using the spur coating machine supplied by JEOL (Japan) Ltd. model 'JFC-1600'.

SEM was conducted according to ASTM E2015-04 (2014) standard using Hitachi Analytical Table Top SEM (bench top) supplied by Hitachi High-Technologies Europe (Germany) GmbH model 'TM-3030'. The images on the samples surface were captured/collected using the field emission gun with accelerating voltage of 5 and 15 kV. Numerous clear microscopic images of the samples fractured surface structure were obtained and discussed.

2.4.2 Light/Optical Microscopy (L/OM)

Light/optical microscopy (L/OM) was conducted according to ASTM E2015-04 (2014) standard using an industrial microscope which was supplied by Nikon (Japan)

Corporation model ‘Eclipse 3 × 2 LV140’. Numerous clear microscopic images of the samples fractured surface structure were obtained and discussed.

3 Results and Discussion

3.1 SEM and L/OM Images of Unmodified and Modified AW Fibre for PLA and PHA Bio-composites Morphological Properties

Figure 1 shows the SEM images of (a) U-AW-PLA and (b) M-AW-PLA, and L/OM images of (c) U-AW-PLA and (d) M-AW-PLA bio-composites. Figure 1a, c showed a big void between the fiber and polymer. It could be observed that the U-AW has a smooth surface structure, and the PLA structure is brittle. Impurities influences the properties, where oily and waxy structure of U-AW fiber causes poor adhesion and absorbance between PLA matrix and U-AW fibre (Mylsamy and Rajendran 2011; Olakanmi et al. 2016). During the quenching process of the PLA bio-composite samples, implosion occurs which made the polymer stronger and closely pack the

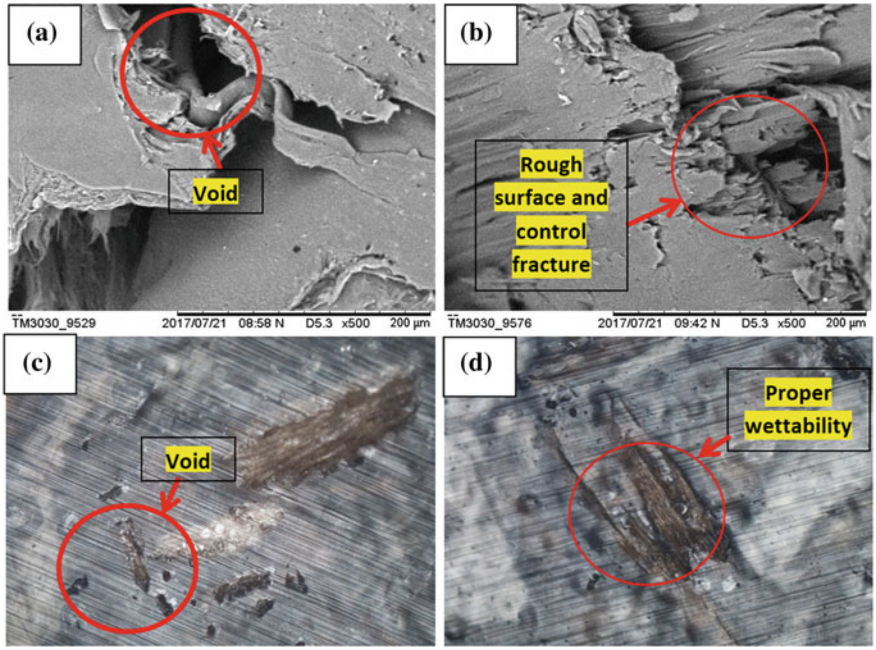

Fig. 1 SEM images of **a** U-AW-PLA and **b** M-AW-PLA, and L/OM images of **c** U-AW-PLA and **d** M-AW-PLA bio-composites

components with each other, hence yielding a brittle structure. Avila-Orta et al. (2013) and Pickering et al. (2016) reported that the controlled temperature upon heating and cooling could influence the structure to become brittle or ductile, depending on the chemical composition reaction and behavior towards temperature change.

Due to the lack of impurities such as wax, oil, some sections of weak lignin, hemicellulose and cellulose of the M-AW fiber, Fig. 1b, d exhibited strong adhesion between fiber and polymer (Olakanmi et al. 2016). Hence, producing a rougher surface, with more pores and stronger micro-fibril branch structures. The desirable adhesion and improved strength in the bio-composites was contributed to the improved interlocking between fiber and polymer (Balakrishna et al. 2013). Chemical modification of the fiber produces desirable wettability and absorption, hence contribute towards improving the adhesion between fiber and polymer. The rough surface, pores and micro-fibril branch structures also produce a higher friction, thus grasp and control the movement of fiber and polymer upon fracture, break up, load or stress applied. Boynard and D’almeida (2000) reported that the controlled movement supports in slowing down the failure movement of the bio-composites.

Figure 2 shows the SEM images of (a), (b) U-AW-PHA and (c), (d) M-AW-PHA, and L/OM images of (e) U-AW-PHA and (f) M-AW-PHA bio-composites. Figure 2a, e shows a void between fiber and polymer and was observed that the U-AW and PHA structure is smooth and ductile, while Fig. 2b exhibited the de-bonding between fiber and polymer. Impurities such as the waxy and oily structure of AW fiber influences the properties, hence caused poor adhesion and absorbance between U-AW fiber and PHA (Mylsamy and Rajendran 2011; Olakanmi et al. 2016). The quenching process of the PHA bio-composites sample caused an implosion to occur, hence produces a stronger polymer with ductile structures with closely pack components. Avila-Orta et al. (2013) and Pickering et al. (2016) reported that the controlled temperature upon heating and cooling could influence the structure to become brittle or ductile, depending on the chemical composition reaction and behavior towards temperature change. Figure 2c shows the ductility phase of PHA bio-composites.

Due to the removal of impurities, such as wax, oil, some section of weak lignin, hemicellulose and cellulose of the M-AW fiber Fig. 2d, f exhibited strong adhesion between M-AW fiber and PHA polymer (Olakanmi et al. 2016). This produces a rougher surface, with more pores and stronger micro-fibril branch structures. The desirable adhesion and improved strength in the bio-composites was contributed to the improved interlocking between fiber and polymer (Balakrishna et al. 2013). Chemical modification of the fiber produce desirable wettability and absorption, hence contribute towards improving the adhesion between fiber and polymer. The rough surface, pores and micro-fibril branch structures also produce a higher friction, thus grasp and control the movement of fiber and polymer upon fracture, break up, load or stress applied. Boynard and D’almeida (2000) reported that the controlled movement supports in slowing down the failure movement of the bio-composites.

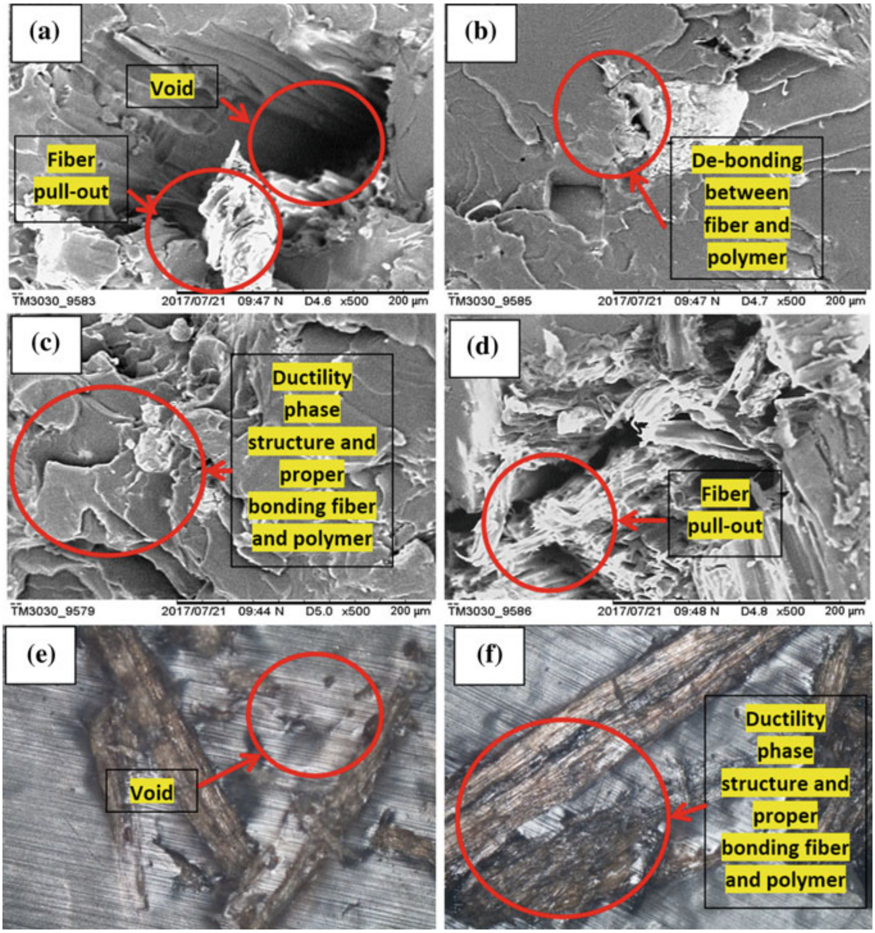

Fig. 2 SEM images of **a**, **b** U-AW-PHA and **c**, **d** M-AW-PHA, and L/OM images of **e** U-AW-PHA and **f** M-AW-PHA bio-composites

3.2 SEM and L/OM Images of Unmodified and Modified Fibre for Polymer Blend (PLA/PHA) Bio-composites Morphological Properties

Figure 3 shows the SEM images of (a), (b) U-AW-PLA/PHA and (c), (d), (e) M-AW-PLA/PHA, and L/OM images of (f), (g) M-AW-PLA/PHA bio-composites. Figure 3a, b exhibited the impurities on the surface of the U-AW fibers and the de-bonding between fiber and polymer, hence producing poor adhesion between fiber and polymer. Weak micro-fibrils branches were observed on the surface, the weak micro-fibrils break easily, which causes improper adhesion between fiber and polymer (Singha et al. 2013). Quenching process for the PLA/PHA bio-composite

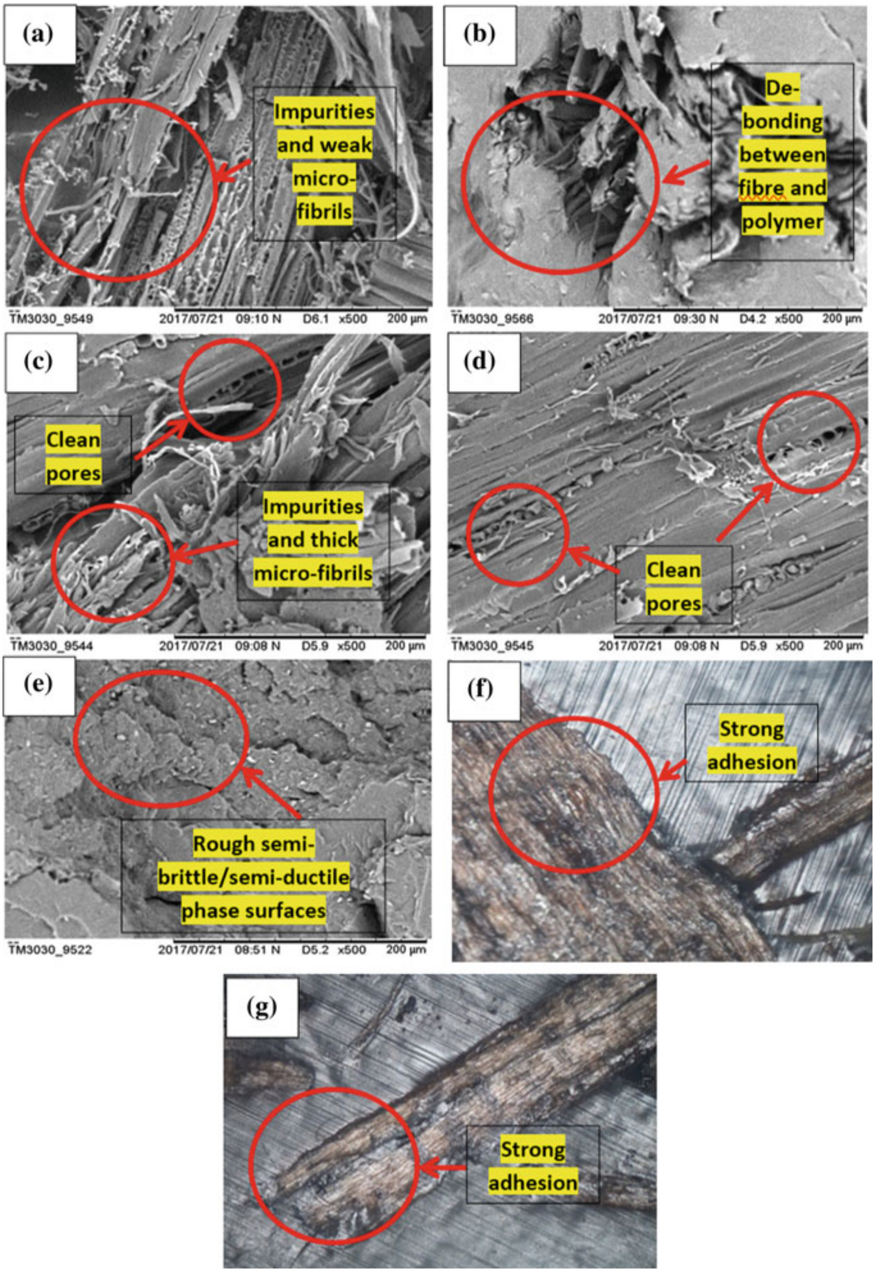

Fig. 3 SEM images of **a**, **b** U-AW-PLA/PHA and **c**–**e** M-AW-PLA/PHA, and L/OM images of **f**, **g** M-AW-PLA/PHA bio-composites

samples caused an implosion to occur, hence produces a stronger polymer with ductile structures with closely pack components. Avila-Orta et al. (2013) and Pickering et al. (2016) supported this, as they reported that the controlled temperature upon heating and cooling could influence the structure to become brittle or ductile, depending on the chemical composition reaction and behavior towards temperature change. Figure 3e shows the rough surface of semi-ductile/semi-brittle interlocking phase of the (PLA/PHA) bio-composites. The interlocking of PLA/PHA were observed in Fig. 3e, hence produces desirable properties as compare to utilizing PLA and PHA respectively as single separate polymer (Dagnon et al. 2009; Ero-Phillips et al. 2012; Li and Mc Carthy 1999; Wagner et al. 2014).

Figure 3c, d, g exhibited that the M-AW fibers have clean pores and thick micro-fibrils branch structures. Polymer could easily be imbedded in the fibers with cleans pores. Thick micro-fibrils branch structures grips better and produces a stronger support, which allows better control over movements and optimizing the load or stress transfer throughout the bio-composites (Olakanmi et al. 2016). Due to the lack of impurities, wax, oil, some section of weak lignin, hemicellulose and cellulose of the AW fiber, Fig. 3f, g exhibited strong adhesion between AW fiber and PLA/PHA polymer. This produces a rougher surface, with more pores and stronger micro-fibril branch structures. The desirable adhesion and improved strength in the bio-composites was contributed to the improved interlocking between fiber and polymer (Balakrishna et al. 2013). Chemical modification of the fiber produces desirable wettability and absorption, hence contribute towards improving the adhesion between fiber and polymer. The rough surface, pores and micro-fibril branch structures also produce a higher friction, thus grasp and control the movement of fiber and polymer upon fracture, break up, load or stress applied. Boynard and D'almeida (2000) reported that the controlled movement supports in slowing down the failure movement of the bio-composites.

3.3 SEM and L/OM Images of Unmodified and Modified Fibre with Nano-enhancement for Polymer Blend (PLA/PHA) Bio-composites Morphological Properties

Figure 4 shows the SEM images of (a) U-AW-NCHB-PLA/PHA, and (b), (c) M-AW-NCHB-PLA/PHA, and L/OM images of (d) M-AW-NCHB-PLA/PHA bio-composites. Figure 4a shows a void between fiber and polymer. It could be observed from the SEM images that the U-AW and NCHB-PLA/PHA has a smooth and semi-brittle/semi-ductile structure. The resulting properties could be caused by impurities such as the oily and waxy structure of U-AW fiber, hence affecting the composite sample by having poor adhesion and absorbance between U-AW fiber and NCHB-PLA/PHA. During the quenching process, it was observed that the NCHB-PLA/PHA samples implode, hence making the polymer stronger with a semi-ductile/semi-brittle interlocking structure closely pack with each other. This statement could be supported

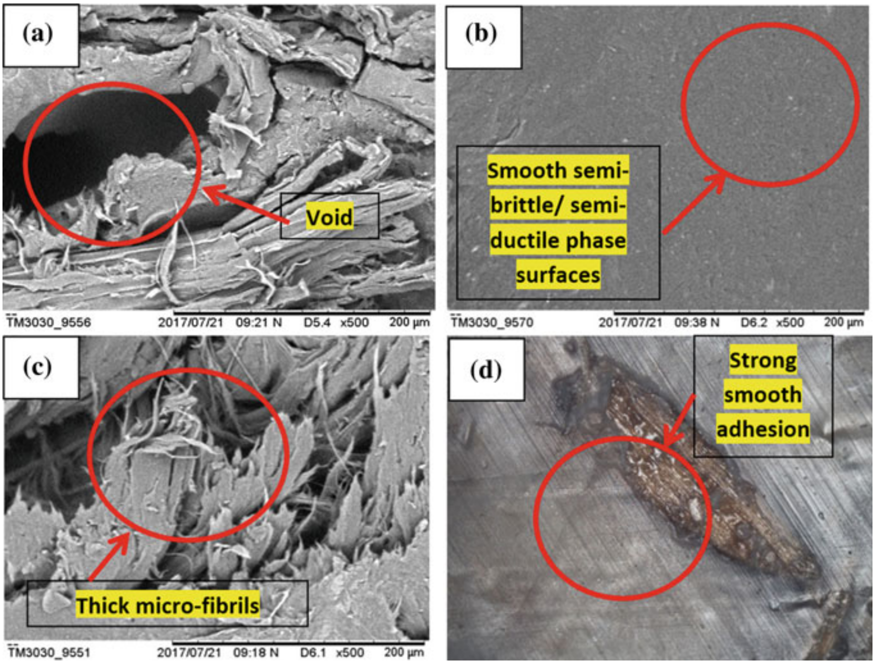

Fig. 4 SEM images of **a** U-AW-NCHB-PLA/PHA, and **b**, **c** M-AW-NCHB-PLA/PHA, and L/OM images of **d** M-AW-NCHB-PLA/PHA bio-composites

some studies by Avila-Orta et al. (2013) and Pickering et al. (2016), where a controlled temperature during heating and cooling could influence a polymer composite to have a brittle or ductile structure, depending on the chemical composition reaction and behavior towards the temperature changes. Due to the fine size of NCHB, it as influences the surfaces of the bio-composites to become smooth. NCHB has a good dispersion in the matrix, hence reducing the development of voids in the bio-composites (Bensalah et al. 2017; Dewan et al. 2013, Hamidi et al. 2008; Haq et al. 2009). Figure 4b shows the smooth semi-brittle/semi-ductile interlocked phase of NCHB-PLA/PHA bio-composites.

As an effect of the removal of impurities such as; wax, oil, some section of weak lignin, hemicellulose and cellulose of the AW fiber, Fig. 4c, d exhibited strong adhesion between M-AW fiber and NCHB-PLA/PHA polymer. This produces a rougher surface, with more pores and stronger micro-fibril branch structures. The desirable adhesion and improved strength in the bio-composites was contributed to the improved interlocking between fiber and polymer (Balakrishna et al. 2013). Chemical modification of the fiber produces desirable wettability and absorption, hence contribute towards improving the adhesion between fiber and polymer. The rough surface, pores and micro-fibril branch structures also produce a higher friction, thus grasp and control the movement of fiber and polymer upon fracture, break up, load or stress applied. Boynard and D'almeida (2000) reported that the controlled movement supports in slowing down the failure movement of the bio-composites.

4 Conclusion

In this chapter, it was discussed that due to the addition of AW fiber, it could influence change in the morphological properties (structure) of the bio-composites. Results exhibited that the U-AW fiber has poor fiber to polymer adhesion in the bio-composites. Chemical modification cause M-AW fiber to be rough, have clean pores and develop more micro-fibrils branch structure. This effect of the chemical modification to the fiber could be due to the removal of lignin, hemicellulose, lignin, wax, oil and other impurities. The chemical modification conducted enables the surface to have more wettability and greater grip, hence producing stronger bio-composites. The PLA/PHA polymer blend produces a rough interlocking structure between PLA and PHA polymer, hence producing stronger and flexible properties in comparison to purely PLA or PHA as a separate polymer. The addition of a Nano-enhancement (NCHB) affects the composite structure by influencing the interlocking structure between PLA and PHA polymers to become smoother. This affect could be caused by the Nano-size of NCHB and being well dispersed during the composite fabrication process. Thus, as a result, reducing voids and improving the properties of the bio-composites.

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Infrared Spectral Functional Group and Thermal Properties of Acacia Wood Bio-composites

M. K. B. Bakri, Md Rezaur Rahman, S. Hamdan, P. L. Nyuk Khui, E. Jayamani and A. Kakar

Abstract This chapter discover spectral functional group and thermal properties of acacia wood bio-composites. A small peak at 2897.08 cm^{-1} for U-AW fiber and 2900.94 cm^{-1} for M-AW fiber was attributed to the C-H stretching and O-H stretching bond structure that contained functional group of alkanes (cellulose and lignin) and carboxylic acids. The small peak in the region of the C-H stretching bond structure can also include a functional group of methyl (CH3), methylene (CH2), and aliphatic saturated (CH). The peak at 607.58 cm^{-1} and 592.15 cm^{-1} for U-AW fiber, and 605.65 cm^{-1}, 559.36 cm^{-1}, 493.78 cm^{-1} and 472.56 cm^{-1} for M-AW fiber is characterized as the =CH bending bond structure from the functional group of alkenes (lignin). Few additional peaks in M-AW fiber spectrum were due to the esterification, which promotes additional free hydroxyl structure, which improves the AW fiber structure for better adhesion with polymer. The peak at 943.19 cm^{-1} and 727.16 cm^{-1} for PLA/PHA, and 875.68 cm^{-1} and 719.45 cm^{-1} for NCHB-PLA/PHA is characterized as the C-H "oop" bond structure from the functional group of aromatics. The peak at 480.28 cm^{-1} for NCHB-PLA/PHA is characterized as the =CH bending bond structure from the functional group of alkenes. The DSC result prove that the reduction in the crystallinity values was an indicator of improvement in the adhesion between fiber and polymer in the bio-composites. The TGA result demonstrated that M-AW-PLA/PHA blend showed two degradation steps. The first step was due to decomposition of hemicellulose, and lignin, weak PHA and PLA bonding and the second step was degradation of cellulose, strong PLA and PHA and other bonding in the polymer blend.

M. K. B. Bakri · Md R. Rahman (✉) · S. Hamdan · P. L. Nyuk Khui
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

E. Jayamani
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

Md R. Rahman (ed.), *Acacia Wood Bio-composites*, Engineering Materials,
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1 Introduction

The bio-composite materials mechanical performances strongly depend on the fiber's orientation and characteristic, the type of polymer, and the quality of adhesion between the two or more components. Most of the wood and natural fiber used are less desirable than synthetic fiber as reinforcement due to the poor compatibility between fiber and polymer. The composition of wood and natural fiber includes mainly cellulose, hemicellulose and lignin. For Acacia wood, Anhydro-D-glucose is the elementary unit of cellulose macromolecule, which contains three hydroxyl (–OH) groups. These large groups of –OH contained in Acacia wood fiber, made it hydrophilic in nature. Most of the Acacia wood and natural fibers are covered with pectin, waxy and oily substances, which make it less attractive as reinforcement with polymer.

The infrared spectral analysis is done by using Fourier transforms infrared (FTIR) spectroscopy. FTIR is used to identify the functional groups of the fiber structure effectively. FTIR tools projected an infrared radiation by creating a spectrum of absorption when targeted onto the samples. This caused the functional groups to vibrate and generate its own data that later were analyzed and grouped accordingly. The sample absorbed the infrared radiation at specific frequencies depending on its types of bonds and functional groups. The molecular fingerprint presented in the frequency graph can either interpreted in absorption or transmission. The vibration frequency created peaks which aid in generating the fingerprint of the molecule. Kabir et al. (2012) stated that the graph peaks help to identify the fiber molecular structure.

FTIR is divided into 3 types of range; far infrared, mid infrared and near infrared. The range of bio-composite FTIR spectrum usually falls in the mid infrared range of 4000–400 cm^{-1} and divided into 4 regions. Stuart (2004) stated that the functional group's presence can be identified depending on its region. There are four regions known according to Stuart (2004); ranging from 4000 to 2500 cm^{-1} (X–H stretching region, may involve O–H, C–H and N–H stretching), ranging from 2500 to 2000 cm^{-1} (triple-bond region), ranging from 2000 to 1500 cm^{-1} (double bond region), and ranging from 1500 to 600 cm^{-1} (fingerprint region).

Referred as the double bond region, the principle band region was due to the C=O and C=C stretching. The C=C double bond groups are identified as the alkenes group. High force on the bond helps to identify triple bond stretching absorption. The absorption peak usually happened in the region between 2300 and 2200 cm^{-1} for C≡N bond and at region 2300–2050 cm^{-1} for C≡C bond. While for C–H stretching bond, the vibration band is used to identify methylene or methyl group of the test sample.

Most of the FTIR for natural fiber composites showed various aliphatic hydrocarbon bands. There is no aromatic ring in any of those aliphatic hydrocarbons.

According to Smith (1998), alkynes, alkenes, and alkanes are three common hydrocarbons groups. Amongst the hydrocarbon, alkanes are the simplest type. It contained C–C bond and C–H bond, molecular groups. While, alkenes contained C=C bond, the molecular groups known as olefins. Lastly, alkynes contained C≡C bond, a molecular group. Thus, this showed that fiber and matrix interaction play a big role in enhanced the natural fiber composites.

TG or DSC analysis has been used extensively to verify the thermal characterization of the materials (Luz et al. 2008) the analytical method is done to understand the thermal stability and structure-property of the bio-composites. Thermal analysis was also used to determine the volatile and moisturizing components, which usually have deteriorating effect on the properties (Nair et al. 2001). It is important to understand the development of composites in both of its service and manufacturing (compression molding, injection molding, extrusion, etc.). It is also known that the structural constituents such as lignin, hemicellulose, and cellulose were sensitive at a set range of temperature. Whereas, the polymer had optimum structural properties at a different range of temperature.

According to Arbelaiz et al. (2006), thermal analysis was used to identify the influence of fiber addition into the polymers. They identified the thermal stability and to find the possibility of degradation during fabrication. It also allowed evaluation of surface modification and fiber content in the matrix. Four stages were identified for degradation processes; moisture evaporation, lignin decomposition, hemicellulose degradation and cellulose degradation. Moisture can exist in free and linked water form. Free water usually attached on the surface of the fiber and evaporated at a lower temperature at the range of 30–150 °C. While, the linked water usually forms with chemical bonds with the hydroxyl groups in lignin and hemicellulose and evaporated at a higher temperature, in the range of 150–500 °C (Randriamanantena et al. 2009). The degradation of lignin, hemicellulose and cellulose begin after the removal of the linked water (Kim and Eom 2001). Pashaei et al. (2011) stated that during fabrication of the composites, under influence of heat, the mode of decomposition and thermal degradation is highly recommended for optimization process of the parameters. TGA curves sometimes have two mass loss steps. All-natural fibers that were associated with water usually had mass loss up to 100 °C due to moisture vaporization that is present in the fiber (Arbelaiz et al. 2006). Due to the hydrophilic nature of the fibers, the total elimination of the water is difficult, which due to the presence of structural bound water molecules. Compared with unmodified fiber, the modified fibers were observed to have less water loss as it has lower hygroscopicity. Considerable mass loss on the fiber due to decomposition of both hemicellulose and cellulose occurred at temperature 330 °C and above. De Paiva and Frollini (2006) stated that above 400 °C, the proto-lignin of the fiber broke.

Brebu and Vasile (2010) stated that at the range of 200–500 °C, the lignin structure decomposed slower than hemicellulose and cellulose structure. Mohanty and Nayak (2010) analyzed the thermal stability of modified and unmodified bamboo/HDPE composites, it showed that the initial decomposition temperature was shifted to the higher temperature of 356 °C for modified bamboo compared with unmodified bamboo 210 °C and the HDPE resin at 348 °C. Furthermore, the modified initial decomposition was shifted to 370 °C compared with raw fiber decomposition of 356 °C. Tajvidi and Takemura (2009) investigated also done numerous of investigation on natural fiber reinforced polypropylene composites. They found that the decomposition of kenaf was very slow compared with other fibers due to the high cellulose content. It is found that the maximum temperature was at 356 °C was obtained than other fiber. Mohanty et al. (2006) stated that jute/high-density polyethylene composites increased when the structure was modified.

2 Experimental

2.1 Materials

The Acacia wood (AW) from Acacia mangium species was provided by Sarawak Forestry Corporation Sdn. Bhd. (SFCsb), Kuching, Sarawak, Malaysia. The AW fibers lengths are less than 3.5 mm. It has a density range of 420–483 kg/m^3 (in green soaked volume) and 500–600 kg/m^3 (in dry condition) (Logan and Balodis 1982; Peh et al. 1982; Peh and Khoo 1984; Razali and Kuo 1983; Wang et al. 1989). Naturally grown AW has a specific gravity (SG) of 0.56 kg/m^3, while the re-plantation grown AW has a lower SG of 0.40–0.45 kg/m^3 (National Research Council 1983). However, in Borneo, the SG can specifically be obtained around 0.43–0.47 kg/m^3 for 6-years old AW (Sining 1989).

For naturalization modification technique, two types of chemicals were used; acidic and alkaline. The pure acetic acid (CH_3COOH) solution was obtained from Fisher Scientific (UK), Ltd., known by the product name “Acetic Acid, ACROS Organics™”. Powdered sodium bicarbonate ($NaHCO_3$) was obtained from Fisher Scientific (UK), Ltd., known by the product name “Sodium Bicarbonate, ACROS Organics™”. Polyhydroxyalkanoates (PHA) was obtained from Jinan Pu Dong Import and Export Trading (China) Co. Ltd. with chemical abstracts service (CAS) number “718630-59-2”. It had a density of 1.2–1.4 kg/m^3 and contained 90% bio-based carbon according to ASTM D6866-16 (2016) standard. Pure grade powdered PLA was obtained from Shenzhen Esun Industrial (China) Co. Ltd. with product code “Al-1001”. It had 100% bio-based carbon according to the ASTM D6400-12 (2012) standards. It had a density of 1.25 kg/m^3 and melt flow index (MFI) of 10–12 g/10 min. Nano-clay, hydrophilic bentonite (NCHB) were obtained from Sigma-Aldrich (US) Corporation with product code “682659”. It has a lighter brown in color and had a particle size less than 25 μ.

2.2 Fiber Preparation

For unmodified fibre, the AW fibres were washed and rinsed with distilled water. Later, it was dried in an open ventilated oven at 80 °C for 24 h. The ventilated oven model "ECOCELL EC55" equipped by MMM Medcenter Einrichtungen (Germany) GmbH was used in this process.

This process involved the use of two different man-made bio-based chemicals. The AW fibres were soaked for 24 h inside 1.5 L of 5 wt% of acetic acid solution (CH_3COOH) mixed with distilled water. After that, 90 g of sodium bicarbonate ($NaHCO_3$) was poured into the soaked AW for 24 h. The reactions of all possible chemical are as below:

$$Na_2CO_3 + 2CH_3COOH \rightarrow 2CH_3COONa + H_2O + CO_2 \tag{1}$$

$$Na_2HCO_3 + CH_3COOH \rightarrow NaCH_3COO + H_2O + CO_2 \tag{2}$$

$$\text{Wood-OH} + CH_3COONa \rightarrow \text{Wood-O-NaH} + CH_3COO \tag{3}$$

The short AW fibres were filtered and rinsed using distilled water to neutralised remaining chemically attached to the AW fibres. Lastly, it was dried in an open ventilated oven at 80 °C for 24 h. All materials obtained were concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.3 Fabrication of the Bio-composites

Pure PLA, PHA, PLA/PHA (1:1 ratio of PLA/PHA), and NCHB-PLA/PHA (1 wt% of NCHB; 1:1 ratio of PLA/PHA) with addition of different fiber loadings (i.e. 5, 10, 15 and 20 wt%) are fabricated as shown in Table 4. The mixtures of the ingredients were done using a drying mixer machine. It was mix blend for about 5 min. The molds used for the bio-composites fabrication, were designed according to ASTM D638-14 (2014) for tensile strength, ASTM D790-17 (2017) for flexural strength, ASTM D4812-11 (2011b) for impact strength, and ASTM D150-11 (2011) for dielectric properties. The molds were cleaned and waxed before they were filled in with the mixture of composite materials (i.e. fiber, polymer and nano-enhancement). Waxes were applied on the molds. The releasing wax contained carnauba that prevents the sample sticks and stuck. Aluminum foils were placed in between the plates, which made the samples easier to be removed from the molds and reduced the formation of bubbles in the samples. The mold was compressed using a hot press hydraulic machine model 'LS-22071' equipped by Lotus Scientific (Malaysia) Sdn. Bhd. under pressure of 1000 psi (6.89 MPa). It was then placed in the hot press machine and heated at 150 °C for 6 min. The samples were then quenched in water at room temperature, according to ASTM E41-92 (2010). The samples were then dried using a hair dryer and were kept concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.4 Methods

2.4.1 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transforms infrared spectroscopy (FTIR) was done based on qualitative and quantitative analysis according to ASTM E168-16 (2016) and ASTM E1252-98 (2013) standards using FTIR Spectroscopy by Shimadzu (Japan) Corporation model ‘IRAffinity-1’. The spectrum scanning was done in the range of wavenumber from 4000 and 400 cm^{-1} for each sample. FTIR used infrared spectrum of emission and absorption of the samples to create unique molecular fingerprint spectrum. The test was repeated a few times for each sample and most representative results were used.

2.4.2 Thermo-Gravimetric Analysis (TGA)

Thermo-gravimetric analysis (TGA) was done according to ASTM E1868-10 (2015) and ASTM E1131-08 (2014) standards using the thermal-gravimetric spectrometer supplied by Shimadzu (Japan) Corp. model ‘TA-60WS’ workstation analyzer. The sample was heated at the rate of 10 °C/min under flowing nitrogen (80 mL/min) between 30 and 400 °C.

2.4.3 Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry (DSC) analysis was done according to ASTM D3418-15 (2015) and ASTM E1269-11 (2011) standards using DSC spectrometer supplied by Perkin Elmer (USA) Corporation model ‘DSC8500 PerkinElmer Smartscan™ Instrument’. The samples were examined under flowing nitrogen (20 mL/min) between 0 and 200 °C. The degree of crystallinity formula is calculated as below:

$$Xc = \frac{\Delta H_m}{w \Delta H_m^0} \times 100 \tag{4}$$

where Xc is the degree of crystallisation, ΔH_m is enthalpy of fusion, w is the weight fraction of polymeric matrix in the composites, and $\Delta H_m^0 = 93.6$ J/g (for the 100% PLA) and/or 109 J/g (for 100% PHA) (Dagnon et al. 2009; Ero-Phillips et al. 2012; Li and Mc Carthy 1999; Wagner et al. 2014).

3 Results and Discussion

3.1 FTIR of Unmodified AW Fibre Spectral Properties

Figure 1a, b show the FTIR spectrum of U-AW fiber and M-AW fiber. Table 1 shows the FTIR spectrum extracted from Fig. 1. The compositions of AW fiber mainly include cellulose, hemicelluloses and lignin (Centikol et al. 2012; Chen et al. 2010; Pandey 1999). According to Khan and Ahmad (1996), Anhydro-D-glucose is one of the elementary units for cellulose macromolecule, which contains hydroxyl groups. This hydroxyl formed intermolecular and intramolecular hydrogen bonds into the cellulose molecules that are responsible for its hydrophilic nature, which cause poor adhesion between fiber and polymer and performance of the bio-composites (Saha et al. 2010; Khan and Ahmad 1996). In Fig. 1, the region of the broad band at 3500–3300 cm^{-1} are characterized as O–H stretching and H-bonded structure that mostly contain functional groups of phenols, alcohols and waters. After chemical modification, this broad band was decreased. This was due to the decrease in the functional group of aliphatic hydroxyls in the fiber, which reacted with chemical treatment to promote free hydroxyls that cause the addition of a peak at 3739.97 cm^{-1}. Addition of free hydroxyl promotes better adhesion between fiber and polymer (Asha et al. 2017; Dwivedi and Mehta 2011; Pickering et al. 2016; Ramadevi et al. 2012). A small peak at 2897.08 cm^{-1} for U-AW fiber and 2900.94 cm^{-1} for M-AW fiber was

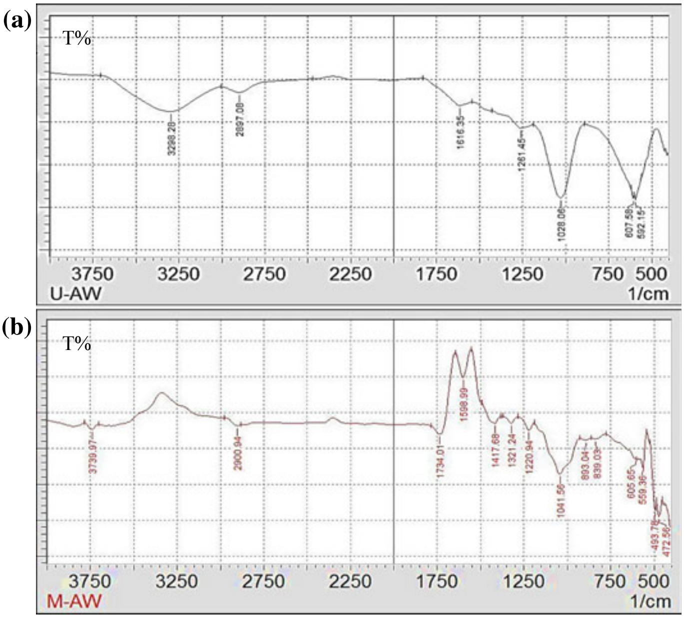

Fig. 1 FTIR spectrum **a** U-AW and **b** M-AW fiber

Table 1 The wavenumber of U-AW and M-AW fibres

Functional group	Wavenumber (cm^{-1})	
	U-AW	M-AW
O–H stretching, Free hydroxyl—Alcohol, Water, Phenols	–	3739.97
O–H stretching, H-Bonded—Alcohol, Water, Phenols	3298.28	–
C–H Stretching, O–H Stretching—Alkanes (CH; CH_2; CH_3), Carboxylic Acids	2897.08	2900.94
C=C Stretching—Alkenes (lignin)	1616.35	1734.01, 1598.99
C–H bending—Alkanes (Cellulose; Hemicellulose; Lignin)	1261.45	1417.68, 1321.24, 1220.94
C–O stretching—Alcohol (Cellulose; Hemi-cellulose; Lignin), Carboxylic Acids, Esters, Ethers	1028.06	1041.56
C–H “oop”—Aromatic (Lignin)	–	893.04, 839.03
=CH bending—Alkenes (Lignin)	607.58, 592.15	605.65, 559.36, 493.78, 472.56

attributed to the C–H stretching and O–H stretching bond structure that contained functional group of alkanes (cellulose and lignin) and carboxylic acids. The small peak in the region of the C–H stretching bond structure can also include a functional group of methyl (CH_3), methylene (CH_2), and aliphatic saturated (CH) (Khalil et al. 2013).

The small peak at 1616.35 cm^{-1} for U-AW fiber, and 1734.01 and 1598.35 cm^{-1} for M-AW fiber is characterized as the C=C stretching bond structure from the functional group of alkenes (lignin) (Owen and Thomas 1989), and the peak at 1261.45 cm^{-1} for U-AW fiber, and 1417.68, 1321.24, 1220.94 cm^{-1} for M-AW fiber is characterized as the C–H bending bond from the functional group of alkanes (cellulose, hemicellulose and lignin) (Owen and Thomas 1989). Slight shift in the peak at 1028.06 cm^{-1} for U-AW fiber, and 1041.56 cm^{-1} for M-AW fiber is characterized as the C–O stretching bond structure from the functional group of alcohol (cellulose, hemi-cellulose and lignin), carboxylic acids, esters and ethers (Hergert 1971; Owen and Thomas 1989). Addition of 893.04 and 839.03 cm^{-1} peak for M-AW fiber is characterized as the C–H “oop” bond structure from the functional group of aromatics (lignin). The peak at 607.58 and 592.15 cm^{-1} for U-AW fiber, and 605.65, 559.36, 493.78 and 472.56 cm^{-1} for M-AW fiber is characterized as the =CH bending bond structure from the functional group of alkenes (lignin). Few additional peaks in M-AW fiber spectrum were due to the esterification, which promotes additional free hydroxyl structure, which improves the AW fiber structure for better adhesion with polymer (Siyamak et al. 2012). It also creates more fibril structures which increased the wettability and improved the adhesion between fiber and polymer.

3.2 FTIR of Nano-Enhancement on Polymer Blend (PLA/PHA) Bio-composites Infrared Spectral Properties

Figure 2a, b shows the FTIR spectrum of PLA/PHA and NCHB-PLA/PHA. Table 2 shows the FTIR spectrum extracted from Fig. 2. A small peak at 2935.66 and 2412.95 cm^{-1} for PLA/PHA, and 2935.66 cm^{-1} for NCHB-PLA/PHA was attributed to the C–H stretching and O–H stretching bond structure that contained functional group of alkanes and carboxylic acids. The small peak in the region of the C–H stretching bond structure can also include a functional group of methyl (CH_3), methylene (CH_2), and aliphatic saturated (CH) (Khalil et al. 2013). The reduction in one of the peaks for NCHB-PLA/PHA was due to the addition of NCHB, which create better interlocking system between amorphous PLA and crystalline PHA (Armentano et al. 2015). This enhanced the adhesion between PLA and PHA.

The small peak at 1730.15 and 1598.99 cm^{-1} for PLA/PHA, and 1730.15 cm^{-1} for NCHB-PLA/PHA is characterized as the C=C stretching bond structure from the functional group of alkenes. This attributed to the mixture of semi-amorphous and semi-crystalline structure of PLA and PHA (Arrieta et al. 2015; Hu et al. 2008; Zhang and Thomas 2011) and peak at 1429.25, 1388.75 and 1247.94 cm^{-1} for PLA/PHA, and 1440.83, 1328.96 and 1247.94 cm^{-1} for NCHB-PLA/PHA are characterized as the C–H bending bond from the functional group of alkanes (Armentano et al. 2015; Arrieta et al. 2015). The peak at 1184.29 and 1099.43 cm^{-1} for PLA/PHA, and 1186.22, 1159.22 and 1097.50 cm^{-1} for NCHB-PLA/PHA is characterized as

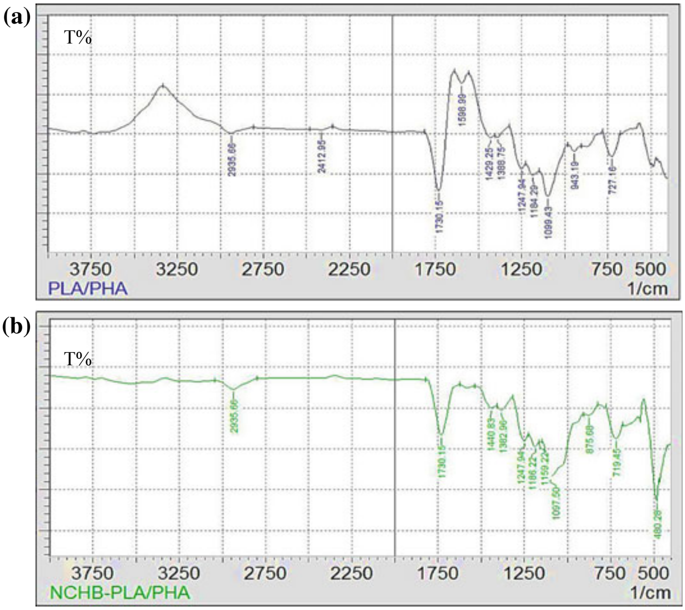

Fig. 2 FTIR spectrum **a** PLA/PHA and **b** NCHB-PLA/PHA

Table 2 The wavenumber of PLA/PHA and NCHB-PLA/PHA

Functional group	Wavenumber (cm^{-1})	
	PLA/PHA	NCHB-PLA/PHA
C–H Stretching, O–H Stretching—Alkanes (CH; CH_2; CH_3), Carboxylic Acids	2935.66, 2412.95	2935.66
C=C Stretching—Alkenes	1730.15, 1598.99	1730.15
C–H bending—Alkanes	1429.25, 1388.75, 1247.94	1440.83, 1328.96, 1247.94,
C–O stretching—Alcohol, Carboxylic Acids, Esters, Ethers	1184.29, 1099.43	1186.22, 1159.22, 1097.50
C–H "oop"—Aromatic	943.19, 727.16	875.68, 719.45
=CH bending—Alkenes	–	480.28

the C–O stretching bond structure from the functional group of alcohol, carboxylic acids, esters and ethers (Armentano et al. 2015). The peak at 943.19 and 727.16 cm^{-1} for PLA/PHA, and 875.68 and 719.45 cm^{-1} for NCHB-PLA/PHA is characterized as the C–H "oop" bond structure from the functional group of aromatics. The peak at 480.28 cm^{-1} for NCHB-PLA/PHA is characterized as the =CH bending bond structure from the functional group of alkenes. Few additional peaks were due to NCHB which promotes additional free hydroxyl structure, which improved the PLA and PHA for better adhesion (Armentano et al. 2015; Hu et al. 2008).

3.3 Thermal Crystallinity Properties of Unmodified and Modified Fiber with PLA and PHA Polymer Bio-composites

Figures 3 and 4 shows the DSC thermogram for U-AW-PLA and M-AW-PLA, U-AW-PHA and M-AW-PHA bio-composites. Table 3 is the numerical representation of the two figures. The degree of crystallization was reduced for modified fiber (M-

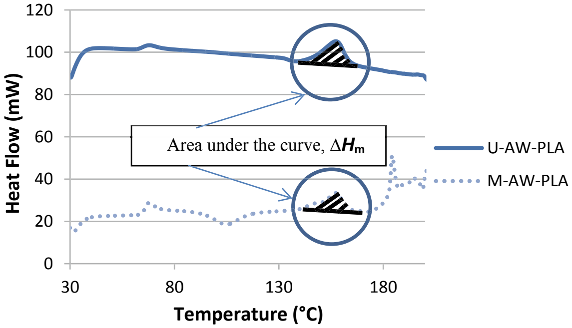

Fig. 3 DSC graph of **a** U-AW-PLA and **b** M-AW-PLA

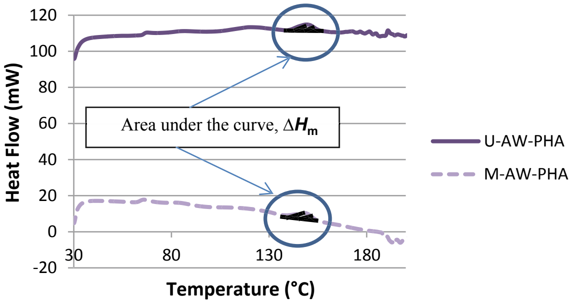

Fig. 4 DSC graph of **a** U-AW-PHA and **b** M-AW-PHA

Table 3 Melting characteristic of U-AW-PLA, M-AW-PLA, U-AW-PHA, and M-AW-PHA

Specimen	Initial melting point (°C)	Final melting point (°C)	Peak melting temperature (°C)	Heat flow—enthalpy of fusion, ΔH_m (Jg^{-1})	Degree of Crystallinity (X_c) (100% PLA)	Degree of Crystallinity (X_c) (100% PHA)
U-AW-PLA	139.6	169.9	158.4	104.7	111.85	96.1
M-AW-PLA	140.9	164.3	157.5	33.3	35.57	30.5
U-AW-PHA	138.3	156.2	150.1	114.7	122.54	105.2
M-AW-PHA	138.8	154.6	148.1	10.3	11.0	9.4

Note $\Delta H_m^0 = 93.6$ J/g (for the 100% PLA) and/or 109 J/g (for 100% PHA) (Dagnon et al. 2009; Ero-Phillips et al. 2012; Li and Mc Carthy 1999; Wagner et al. 2014)

AW-PLA and M-AW-PHA) bio-composites. The reduction in the crystallinity values was an indicator of improvement in the adhesion between fiber and polymer in the bio-composites. The hydrogen bond formation between fiber and polymer is known to reduce the crystallinity values of the bio-composites (Ball et al. 2004; Kumar et al. 2010). It was also noticed that the peak melting point of the bio-composites were higher for unmodified AW fiber compared with the modified AW fiber. This was due to the removal of cellulose, hemicellulose, lignin and other impurities, which improved the interaction between fiber and polymer (Bozkurt et al. 2007; Sinha and Rout 2008). Comparing between PLA and PHA, it was noticed that PHA was slightly more crystalline than PLA. This implied that PHA is more ductile than PLA, even though both are semi-crystalline/semi-amorphous, respectively (Armentano et al. 2015; Wagner et al. 2014). It is known that higher degree of crystallinity implies more brittle structure (Bessell et al. 1975). The higher amount of brittle structure in the polymer contributes to a more desirable mechanical strength of the bio-composites (Armentano et al. 2015).

Fig. 5 DSC graph of **a** U-AW-PLA/PHA and **b** M-AW-PLA/PHA

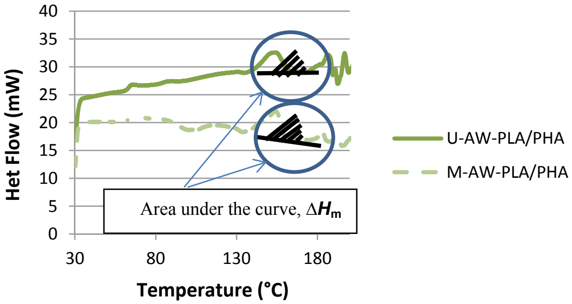

Table 4 Melting characteristic of U-AW-PLA/PHA and M-AW-PLA/PHA

Specimen	Initial melting point (°C)	Final melting point (°C)	Peak melting temperature (°C)	Heat flow – enthalpy of fusion, ΔH_m (Jg^{-1})	Degree of Crystallinity (X_c) (100% PLA)	Degree of Crystallinity (X_c) (100% PHA)
U-AW-PLA/PHA	136.3	163.7	152.4	32.4	34.6	29.7
M-AW-PLA/PHA	133.3	162.5	154.7	21.9	23.4	20.1

Note $\Delta H_m^0 = 93.6$ J/g (for the 100% PLA) and/or 109 J/g (for 100% PHA) (Dagnon et al. 2009; Ero-Phillips et al. 2012; Li and Mc Carthy 1999; Wagner et al. 2014)

Figure 5 shows the DSC thermogram for U-AW-PLA/PHA and M-AW-PLA/PHA bio-composites. Table 4 contains the numerical representation of the graph. The polymer blend caused reduction in the crystallinity values for both PLA and PHA, which proved the interlocking structure between PLA and PHA (Armentano et al. 2015; Wagner et al. 2014) The crystallinity values for the polymer blend were noticed to have reduced when chemically modified AW fiber was used, compared with unmodified AW fiber. This indicates that the modified AW fiber have desirable adhesion with polymer blend. According to Kumar et al. (2010), the hydrogen bond formation between fiber and polymer reduces the crystallinity values of the bio-composites.

3.4 *Thermal Degradation Properties of Bio-composites*

Figure 6 shows the TGA graph for U-AW, M-AW, M-AW-PLA, M-AW-PHA, M-AW-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. It was noticed that the major degradation began at 280 °C. There are three stages of decomposition for AW fiber. The first stage occurred between 40 and 100 °C, which is attributed to evaporation of water. The second stage happened from 230 to 288 °C, which is

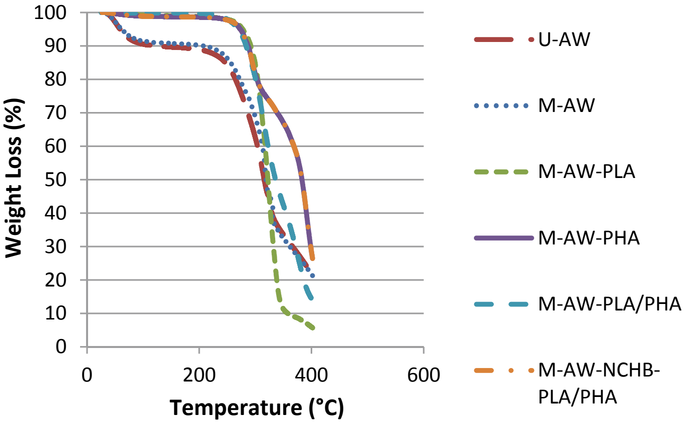

Fig. 6 TGA graph of U-AW, M-AW, M-AW-PLA, M-AW-PHA, M-AW-PLA/PHA, and M-AW-NCHB-PLA/PHA

related to degradation of lignin and hemicellulose. The third stage was from 300 °C and above, which indicated the degradation of cellulose and other cellulosic matters (Morshed et al. 2012; Raghavendra et al. 2014). Modified AW fibers have higher thermal stability than the unmodified AW fibers. The high thermal stability was due to the additional hydroxyl group, which improved interaction between the binder and the fiber, which produces additional intermolecular bonding between fiber and polymer (Mohanty and Nayak 2010). According to Nguyen et al. (1981), the thermal decomposition of cellulose fiber begins at temperature in the range of 210–260 °C.

It was noticed that PLA has higher thermal decomposition than PHA, which was due to the intramolecular and intermolecular trans-esterification that led to the formation of lactide and cylic ligomers and cis-eliminations and led to the formation of acrylic acid and acyclic ligomers (Arrieta et al. 2017; Kopinke et al. 1996). The M-AW-PLA/PHA blend showed two degradation steps. The first step was due to decomposition of hemicellulose, and lignin, weak PHA and PLA bonding and the second step was degradation of cellulose, strong PLA and PHA and other bonding in the polymer blend (Arrieta et al. 2017). The addition of NCHB in the polymer blend had minimal effect on the thermal stability of the polymer blend due to its small amount. Therefore, it only affects the dispersion and adhesion of the bio-composites. It was also noticed that M-AW-PHA and M-AW-NCHB-PLA/PHA bio-composites fall under the same line. The reason behind this behavior could not completely be verified. However, it is speculated that additional carbon atom rearrangement due to addition of NCHB may triggered the similar pattern for the graph.

4 Conclusion

In this chapter, it is noted that the chemical modification on the AW fiber cause changes in the functional group behavior of bio-composites. It was found that free hydroxyl group were created, which promotes better adhesion between fiber and polymer. Chemical modification on the fiber cause M-AW fiber to be rough, clean pores and more micro-fibrils branches structure, which was due to removal of lignin, hemicellulose, lignin, wax, oil and other impurities. This allowed more surface wettability and greater grips, which create stronger bio-composites. Polymer blend (PLA/PHA) creates rough interlocking structure between PLA and PHA polymers, which create stronger and flexible properties than its own polymers. All these modifications influenced the mechanical and morphological properties. Lower crystallinity indicated better adhesion between fiber and polymer. It was noticed that PLA and PHA had semi-amorphous and semi-crystalline structure, respectively. The polymer blend improved the properties and adhesion by lowering down crystallinity structure of the polymer, while NCHB further improved the adhesion. The thermal stability shows that modified AW fiber is stable than the unmodified AW fiber. This was due to additional hydroxyl group; this produced intermolecular bonding between fiber and polymer. PLA was noted to have higher thermal stability than the PHA due to intramolecular and intermolecular trans-esterification. Polymer blend further increased the thermal stability of the polymers. The addition of NCHB had minimal effect on the thermal stability due to the small amount used, however it only affects the dispersion and adhesion of the bio-composites. All this modification influenced the mechanical, morphological and spectral properties.

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Environmental Impact Analysis of Wood and Natural Fiber Bio-Composites

E. Jayamani, H. L. Tan, D. A. Benhur and M. K. B. Bakri

Abstract In this chapter, a brief review on the practicability of using fiber reinforced polymer (FRP) in construction applications, particularly in the reinforced concrete structures were reported. The environmental and performance issues of conventional building materials such as concrete and steel were discussed in detail. The prospect of implementing FRP composite in this area was also discussed. To assess the undesirable environmental burdens of using FRP, the fundamental concept, procedures and several components of LCA have been discussed thoroughly. The existing LCA case studies on different materials were also critically reviewed for better understanding. The effects of the composition variation in the FRP system on the environment were also examined in this chapter. The popularity of LCA particularly in the use of composites in construction industry has been very low, due to the inherent complexities associated with the building itself.

Keywords Wood · Hybrid · Fiber · Life cycle analysis

1 Introduction

1.1 *Effect of Construction and Building Industry to the Environment*

Public concern and awareness of global environmental issues such as resources depletion and climate change have gradually emerged in recent years. In extend human activities may greatly pose environmental problem from one industry to another. While showing rapid expansion and becoming one of the most active industries

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E. Jayamani (✉) · H. L. Tan · D. A. Benhur
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia
e-mail: ejayamani@swinburne.edu.my

M. K. B. Bakri
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia

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in the world, construction and building industry has also been a major energy and material consumers. It brings substantial burdens to the environment throughout its entire life cycle, from the acquisition of raw materials to the end-of-life treatments. For instance, as compared to other economic activities, the construction industry is responsible for about 50% of the world raw material consumption, which is expected to increase even more in coming years (Maxineasa and Ţăranu 2013). Another piece of research by Abd Rashid and Yusoff (2015) also presented that all the constructions around the world represent around 30–40% of the total share for energy usage. Most of the world carbon dioxide (CO_2) emission is also contributed by this industry, generating approximately 50% of the total amount (Khasreen et al. 2009).

In fact, most of these environmental loads is directly contributed by the main component of the construction industry—the building material. For example, in the energy consumption wise, Esin (2007) declared that constructive materials are responsible for approximately 10–15% of the total energy used by the building throughout its service lifetime. To be more specific, the production and processing of building materials are proven to have brought about significant negative effects to the environment, according to Maxineasa and Ţăranu (2013). This can be simply explained by the huge materials and energy usage during the process, which, ultimately, also leads to harmful discharges, emission and unwanted wastes to the environment (Esin 2007). For building 1 m^2 gross floor area of a typical, standard construction in Spain, it is required to combust approximately 150 L or more petroleum to generate enough energy for producing the required amount of building materials, as approximated by Zabalza Bribián et al. (2011). It is notable that some of the most commonly used conventional constructive materials such as steel and cement contribute to high environmental impacts. The production and processing of these two materials contribute significantly to both primary energy demand and CO_2 emission from the total impact of twelve building materials, with steel having the highest contribution of 25.5% for the first parameter and concrete making also the greatest share of 30.3% for the second parameter (Zabalza Bribián et al. 2011). To highlight the severity of this scenario, a piece of research showed that the production of cement alone has contributed for approximately 5–7% of the world total CO_2 emission (Maxineasa and Ţăranu 2013).

Being perceived as one of the biggest challenges of the 21st century, strategies are implemented that optimally unite both development and environment within one single context to achieve the so-called environmental sustainability (Ortiz et al. 2009). According to Zaman et al. (2014), reducing the usage of natural resources, lowering the risks associated with human health, developing sustainable design strategies, improving the performance of the product designed are also among the factors to be considered, while dealing with the sustainability issue.

1.2 Sustainable Development and Sustainability

Fiber reinforced polymer (FRP) is an extremely unique yet versatile group of composites, which has been heavily used in different types of application and industry in recent decades. While being fascinated by the promising prospect of FRP composite in various applications, concern related with the sustainability of this technology still appears to be a big question mark for future researchers. It is popularly known that the raw materials required for fabricating the fibers and matrix are usually derived from one of the depleting non-renewable resources—the fossil fuels. Besides, the manufacturing processes of the fiber components are also very high energy consuming for the fact that high temperature is needed to accomplish the production. FRP composite also exhibits great potential in mitigating the negative environmental effects for some instances. As explained by Zaman et al. (2014), the high durability of FRP composites lengthens the useful life time of a product thus, in the long run, reducing the usage of resources and minimizing the requirement for maintenance. Nevertheless, it is essential to explore deeper into all the aspects and have a wider view before concluding on the future of FRP composites. However, things are clear that the environmental concern of FRP composites remains as the critical barrier from categorizing it as a sustainable material, which, in turn, significantly limits its growth in the engineering field. It is necessary to profoundly assess and evaluate the environmental issues of this material using a proper and systematic approach.

1.3 Life Cycle Assessment (LCA)

When comparing various environmental management tools in the market, LCA has been the most widespread framework or analytical decision-making support tool used in different economical industries to evaluate and examine the potential environmental loads throughout the entire life-cycle of a product system (Khasreen et al. 2009). For certain cases, LCA can also be utilized for comparing the alternative products of similar functionality and identify the most optimum one based on the result of the analysis (Leterrier 2000). A complete LCA, based on the international standards set by ISO 14040/1/2/3 series, comprises of four distinct sequential phases, which are goal and scope definition, Life-Cycle Inventory (LCI), Life-Cycle Impact Assessment (LCIA) and lastly, the interpretation of result (Khasreen et al. 2009).

Life Cycle Assessment (LCA) can be regarded as the best tool that can be applied to effectively optimize the aspects of sustainable development and examine the sustainability concern of the construction industry (Maxineasa and Ţăranu 2013). In various industries and applications, this environmental management tool has been utilized to investigate and assess the environmental impacts of a product, system or process (Khasreen et al. 2009). As defined in ISO 14040/2006 and ISO 14044/2006, LCA is an analytical decision support tool that evaluates the potential environmental loads throughout the entire life-cycle of a product system, starting from acquisition and processing of raw materials, to manufacturing phase, distribution or transportation phase, usage phase, maintenance and finally, end-of-life treatment (Khasreen et al. 2009). It compiles and evaluates the use of resources (inputs) and the environmental emissions or discharges (outputs) of the product system under the consideration before expressing its potential environmental consequences in the numerical form (Maxineasa and Ţăranu 2013). LCA has become increasingly popular, for the aim of delivering a more eco-efficient world.

1.4 Fiber Reinforced Polymer (FRP) Composites

The use of FRP composites in construction sector are very limited to minor building components such as doors, windows, decorations, baths, kitchenware and others. Additional FRP structural parts that can be seen in the market are beams, columns and reinforcing bars (Zaman et al. 2013). It can be observed that initially FRP composites were only used as 'secondary structures', supported by the 'primary structures' which were made up by the conventional building materials (Hollaway 2010). One of the most common applications of FRP in the construction field is the rehabilitation and repair of building parts, for instance, structural strengthening of concrete structures (Zaman et al. 2013). Due to the advantage of FRPs over the traditional methods, the use of these materials as reinforcing component, both internally and externally, in different civil structures has also been popular in recent years, particularly in bridge decks and concrete structure. As stated by Zhang (2014), steel bars in reinforced and stressed concrete structure have also been gradually substituted by FRP composite bars, overcoming the some of the issues associated with metal such as corrosion in chemically aggressive environments. This common phenomenon not only lead to structural instability, but also reduce the durability of the material while increasing the lifecycle cost of the structure itself due to the necessity of maintenance (Richardson and Drew 2011). The application of FRP composites to reinforce concrete structures has significantly increased over the years since it was first investigated in 1960s. Aside from superior longitudinal strength, it also has good fatigue resistance and low density.

The feasibility of FRP reinforcement for concrete structure is further assured by the ease of construction and handling, cost incurred, material durability as well as its suitability to replace materials that hinder the radio-wave propagation in certain applications. The examples of traditional FRP composite constituents that have been used in this application are of various combinations between epoxy, polyester or vinylester polymer matrices with E-glass or carbon fibers as reinforcing phase (Zaman et al. 2013). An experiment carried out by Richardson and Drew (2011) investigated on the feasibility of using FRP rebar in concrete structure reinforcement, by testing and comparing the performance of a glass/epoxy FRP against the traditional steel rebar. The result showed that the concrete structure reinforced by FRP composite is able to withstand high number of loads and its failure mode is generally more desirable since the concrete structure failed steadily rather than breaking instantly. From the perspective of natural fibers, some of them have been proven to be feasible for being used as the reinforcing components in concrete structure, including jute, bamboo, sisal and coir. Possessing outstanding mechanical and chemical resistances, hemp fiber has also attracted the attention from researchers for using it as a new concrete reinforcing material (Awwad et al. 2012). As compared with other natural fiber contenders, hemp fiber does not only outstand due to its structural properties, but also for the fact that using hemp mat in substituting glass mats has been proven to achieve approximately 20% of both cost and weight savings (La Rosa et al. 2013b).

2 LCA Framework

2.1 Procedures of LCA

The very first step of LCA is defining the goal and scope of the intended study clearly, which, according to Khasreen et al. (2009), is a very crucial step that eventually may have significant influence on the LCA result. Based on ISO 14040, this stage covers the information ranging from the application chosen for study, to the purpose of analysis as well as the targeted audience (Ortiz et al. 2009). To be more specific, study holder, must explicitly define the product system of the analysis along with its functions, system boundaries of the study, the functional unit, impact categories chosen, impact assessment methodologies, assumptions made for the sake of simplicity, limitations which include lack of data as well as the data requirement for accomplishing the study. Proper identification of system boundaries is exceptionally important, in which the LCA practitioner must decide on whether the study considered only one phase throughout the product system lifetime rather than the entire cycle, or, in the context of construction, considering only one small system rather than the building. The determination of functional unit, on the other hand, is also a critical step as it serve as a reference through which all the inputs and outputs enter and leave the defined product system (Khasreen et al. 2009). In conducting, the LCA related to building, the most commonly used functional unit can be either one of the followings: m2 floor area, m3, number of occupants and ton of material. Regardless of the variation in the selection of functional unit from one study to another, it is important to know that environmental loads result calculation within one LCA analysis must be consistent with the selected function unit.

With the well-defined goal and scope, Life-Cycle Inventory (LCI), or more popularly known as inventory analysis, can then be proceeded. Being the most intensive step in LCA, LCI collects and compiles an inventory of all the relevant inputs and outputs for each defined life-cycle phase of the product system under the consideration, including the emissions to air, land and water as well as the inflow and outflow of mass and energy. With that in mind, one can note that LCI also plays a key role in a successful LCA as the data collected will serve as the foundation for the entire analysis. Data collection depends highly on both the goal and scope defined previously, as they will determine whether a process or item will be included in the assessment. As for the source of data, it can be one or more of, but not limited to the followings: environmental inventories, laboratory results, data from governmental department or industry, national databases, academic sources and engineering-based judgments. In building sector, however, the LCI typically uses the data banks of building materials and component combinations (BMCC). As defined by ISO standard, data collection is not the only element of inventory analysis, it also involves data calculation and allocation procedures.

The former is important for validation of complied data as well as building the relationship between the data with the unit processes and functional unit. The latter is only essential when the LCA practitioner is analyzing a system consisting of multiple products or otherwise, it may not be necessary. Proper data selection in this stage is also crucial as the quality of the data collected has direct influence on the result accuracy, becoming the deciding factor for the success or failure of the intended analysis. There are also cases where issues like data unavailability might happen during LCI, forcing the need of modifying or redefining the scope, objectives or system boundaries of the study. There are several indicators listed by Khasreen et al. (2009) on which the quality of the data obtained can be assessed, including data reliability, data completeness, temporal correlation, geographical correlation and technological correlation. Among these, geographical factor is deemed to possess highest influence on accurate data collection. Although experts believe that it is essential for all nations to have their own databases based on the resources and conventions in the respective building sector. It is still not very well-established, especially in developing countries, due to lack of data completeness, data transparency, accessibility to LCA databases and absence of internationally accepted methods used for LCA analysis (Khasreen et al. 2009).

In general, LCI can be accomplished by three major approaches, which are process analysis, input/output analysis and the combination of the previous two methods, namely, hybrid analysis. Process analysis, being the most traditional method, assess both the direct (all the main processes) and indirect (transport of materials or equipment, operation of machine and manufacturing of equipment) energy inflows to each individual product process. In this case, the system boundary can be defined in such a way that covers all the inputs where data are readily available, or intentionally neglects the insignificant environmental impacts associated with the system outside the defined boundaries (Zhang 2014). Although process analysis can be comparatively higher in terms of accuracy than input/output analysis, the former approach, however, can be impracticable in some scenarios due to the difficulty in understanding the entire production process of the system, making the analysis to be incomplete (Khasreen et al. 2009). This can be overcome by the latter approach, which is deemed more comprehensive as it uses the national average energy data of each economical industry. Based on this data, the inputs of this analysis encompass energy and natural resources consumed by the system while the output can be any emissions by the system to the environment. However, the data available can be very limited number to only several industries or in some instances, the data required for the process under consideration may have been contained in one aggregated number, which makes it very challenging that LCA practitioner to accurately identify the exact contribution directly incurred by one process from the total (Zhang 2014). With these inevitable issues, hybrid approach has been developed which fully exploits the advantages of both the approaches mentioned previously to overcome their respective shortcomings. In other words, hybrid approach can be said as a combination of both analysis methods, which allows the researchers to compile a data inventory of high quality (Khasreen et al. 2009).

With respect to the LCI results, Life-Cycle Impact Assessment (LCIA), as the third step, will estimate and examine the magnitude and significance of environmental impacts for the modelled product system using the appropriate impact categories and category indicators, based on the definition of ISO standard (Zhang 2014). Multiple steps make up LCIA itself but in general, there are three key elements within this assessment—impact categories selection, classification and characterization. Starting with the first element, the definition and selection of suitable impact categories may vary from one LCA study to another and they depend highly on the objective of the study, significance of the impacts to the environment as well as the availability of data (Khasreen et al. 2009). The popular impact categories within the construction industry are global warming, acidification, abiotic resources, human toxicity, eutrophication and ozone depletion, according to Ortiz et al. (2009). Classification, as the next step, involves assigning the LCI results, such as resources consumed and emissions, to different impact categories accordingly. As an example, the discharge of CO_2 is classified as the cause of global warming. Whereas characterization is important when multiple results in LCI are contributing to the same impact categories, that it converts and combines these results with respect to the relevant impact category based on a conversion factor called equivalency factor. For instance, the impacts of CO_2 and methane (CH_4) on climate change, or more specifically global warming, can be modelled using the relevant equivalency factor, which is greenhouse gases potential (GWP) in this case, to compare the relative severity of each emission toward the similar impact category.

Normalization, grouping and weighting are some of the optional steps in LCA which are basically used to express the impact categories in a non-dimensional manner so that a comparison can be made between each of them easily (Khasreen et al. 2009). In general, the impacts can be assessed based on two general methods, which are mid-points and end-points approaches. The former assesses the environmental effects related to climate change, photochemical oxidation, eutrophication, human toxicity, acidification and others while the latter further divides and assesses these impacts with respect to the damages caused to natural resources, ecosystem and humans (Ortiz et al. 2009). According to Scientific Applications International Corporation (2006), modelling the LCA using mid-point analysis will lower the complexity of the study, thus minimizing the needs of forecasting the effects, choices of value for calculation as well as making additional assumptions, which are all required for end-points analysis.

The final phase of LCA will be the interpretation of result, focusing on analyzing and evaluating the findings from the preceding stages before the conclusion and recommendations can be formulated (Ortiz et al. 2009). Interpreting the LCA result will also provide the LCA practitioners with better understanding on the environmental impacts so that they can compare and decide on which of considered alternatives is the best and most optimum choice (Scientific Applications International Corporation

2006). Apart from those, this step is also aimed to present all the results obtained in a complete, profound and understandable manner, with reference to the goal and scope defined at the initial stage of study (Khasreen et al. 2009).

2.2 LCA in Construction and Building Sector

Since year 1990, the use of LCA has been expanding slowly in construction industry due to its importance in assessing the environmental impacts caused by the buildings (Khasreen et al. 2009). According to Ortiz et al. (2009), there are generally two major methods of applying LCA in this industry, namely Building Material and Component Combination (BMCC) and Whole Process of Construction (WPC). The first method, which is used in most of the LCA case studies, focuses on assessing the building parts, components and material. In WPC, the object analysed is the entire building itself, whereby the analysis will be carried out on all the materials and processes involved in constructing a building rather than just focusing on one single product. There is a number of factors that make WPC to be a comparatively more complex approach in this context. For instance, as emphasized by Khasreen et al. (2009), it is very hard to accurately anticipate all the possible processes involved in the entire life-cycle of a building for its long lifespan, which is usually assumed to be 50 years or more. Modifications can also be made onto the form, design as well as the function of the building occasionally during its use phase. While these are very difficult to be foreseen, the environmental impacts of the building contributed by these changes can hardly be predicted as well, which severely lower the accuracy of the entire LCA because study actually showed that majority of the environment impacts of a building come from this life-cycle phase. Also, since there is no standardization on how a building should be designed, the uniqueness of each building will further increase the complexity of WPC as a completely new study has to be conducted based on specific scenario. The involvement of various stakeholders in the construction industries such as designers, developers, material manufacturers, users and others also make it very difficult to predict the life-cycle processes (Khasreen et al. 2009).

2.3 Existing LCA Case Studies

La Rosa et al. (2013b) used ReCipe impact assessment method in conducting LCA on a product made up by hybrid glass-hemp-fiber/thermoset composite based on 17 impact categories. The functional unit defined was the seawater pipeline elbow fitting with 20 years of useful lifetime. The system boundaries of the study covered all the phases from material acquisition to the production of the fitting, transportation, use phase and end-of-life phase. In this analysis, it was assumed that the cultivation of organic hemp fiber does not require fertilizers and pesticides while the effect of hemp cultivation on the global warming was also negligible considering that the natural fiber is carbon neutral. The result showed that the inclusion of hemp fiber reduces the use of both glass fiber and epoxy resin and hence, the environmental impact.

Another LCA was conducted by La Rosa et al. (2013a) which examined on the environmental effects of an automotive interior side panel based on 11 impacts categories, using SimaPro 7.2 software and, again, ReCipe impact assessment methods. The functional unit used was the composite panels with 10 years of useful life, made up by different materials, with one of which being the plant-based hemp-fiber/epoxy composite while another being the petroleum-based glass/epoxy composite. In terms of system boundaries, the analysis considered all the processes in the panel production phase, transportation of materials, use phase as well as the end-of-life phase. The assumption made on the hemp cultivation in previous study was also applied in this study. It was concluded that the use of plant-based materials allows significant environmental impact reduction, especially in production and use phases, mainly due to the weight saving benefit provided by these materials.

Using SimaPro software and Eco-indicator 99 impact assessment method, Xu et al. (2008) compared the environmental effects of two structures having the same size (127 mm × 127 mm × 6 mm), with one of which entirely made up by polypropylene while the other one is a three-layer-sandwiched composite structure with the wood-fiber sheet at the middle covered by two polypropylene sheets. The system boundaries defined for this study considered only the production and processing of the raw materials as well as the manufacturing phase of the composite layer. This study proposed the concept of 'material service density' that referred to the volume of material needed to meet a specific application or strength requirement. In other words, it emphasized the importance of comparing the structures with respect to similar structural performance, which is tensile strength in this case. The result concluded that the wood-fiber/polypropylene composite clearly stands out in the environmental perspective although the use of chemical products during the fiber cultivation process may lead to negative impact to the natural environment. Also, the dominant factor of environmental impacts is found out to be contributed by the polymer matrix, which is the polypropylene resin in this study.

Similar finding is also presented in another LCA which compared ten impact categories on two sandwich structures of similar size (0.4 m × 0.4 m × 0.02 m) and thermal properties, made up by hemp-fiber/bio-epoxy resin and glass/petroleum-based-epoxy resin respectively, using SimaPro 7.2 software and CML 2000 v2.0 (La Rosa et al. 2014). The system boundaries considered in this study was cradle-to-gate, which covered phases from the acquisition and processing of raw materials to the production phase of the sandwich panels only. Although the use of bio-based composite is proven to have higher environmental standing, the use of epoxy resin contributed for more than 85% of the total environmental impact by the sandwich structure. The limitation of this analysis was also highlighted that the researchers ignored some of the important criteria for process and material selection, such as the efficiency of the process as well as the properties and abundancy of the chosen materials.

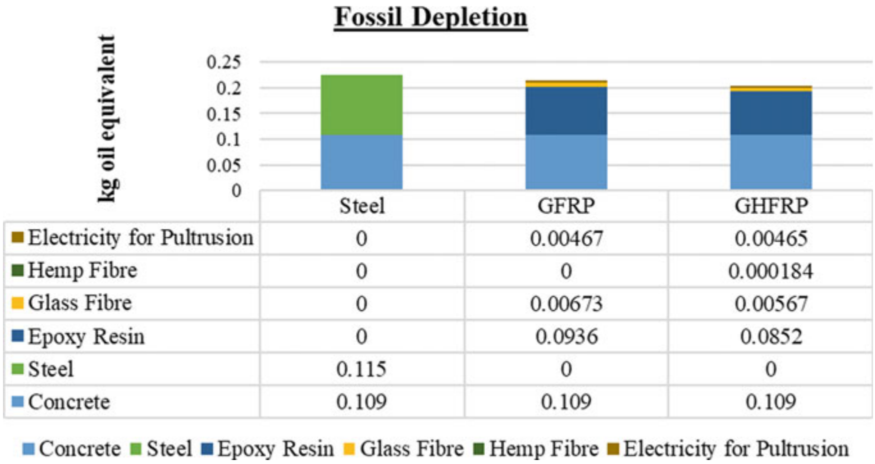
LCA carried out by Corbière-Nicollier et al. (2001) analyzed the environmental impacts contributed by two types of standard transport pallets with similar service requirements for 5 years of useful lifetime, based on CST 95, CML 92, Ecopoints and Eco-indicator 95 impact assessment methods. Using the same polymer matrix (polypropylene), the researchers compared the effects of using glass fiber and natural China reed (CR) fiber in the functional unit defined. The system boundaries chosen for this study covered all the processes from production phase, to use phase and end-of-life treatments. Similar results were also obtained in the LCA that the use of natural FRP is comparatively more environmental-friendlier. In addition, seeing how the matrix phase contributes significantly in the overall environmental impacts, the researchers suggested that other natural or biodegradable materials could be used for replacing the original petroleum-based polymer materials to further improve the environmental standing of natural FRP.

3 Steel RC Beam System Versus Glass-Fiber/Epoxy and Glass-Hemp-Fiber/Epoxy RC Beam System

A comparative analysis has been performed to examine the environmental advantages or disadvantages of substituting the reinforcing steel in the conventional concrete beam structure using the proposed FRPs, which were anticipated to be a more ecological approach in material design. Result demonstrated that the use of GFRP (Glass fiber reinforced polymer) as the reinforcing component leaded to direct reduction in negative environmental impacts, particularly in fossil depletion and climate change. The modification made allowed a reduction of approximately 4.5 and 8% respectively. On the contrary, the use of GFRP rebars caused an increase in water depletion by around 20%.

3.1 Fossil Depletion

Scrutinizing on the steel reinforced concrete beam, Fig. 1 shows that the concrete and steel rebar contributed an equally high impact on the fossil depletion. Both materials, as proven by the LCA study conducted by Zabalza Bribián et al. (2011), required a huge amount of energy demand during the production phase, especially the heat energy. The high value of fossil depletion in this beam structure can be easily linked to the requirement of generating sufficiently high heat energy to cater the demand of the heating processes during the fabrication of these materials. Usually, the energy generation consumes and combusts the non-renewable energy resource, which is particularly the fossil fuel (Burchart-Korol 2013). From here, it can be deduced that the higher amount of concrete and steel rebar used in the structure, more energy required to manufacture the material, the greater amount of fossil fuel will be consumed in the process and hence, the higher value of fossil depletion. Considering that concrete was the main material of a beam structure, this study will only focus on replacing the reinforcing component, which was the steel rebar.



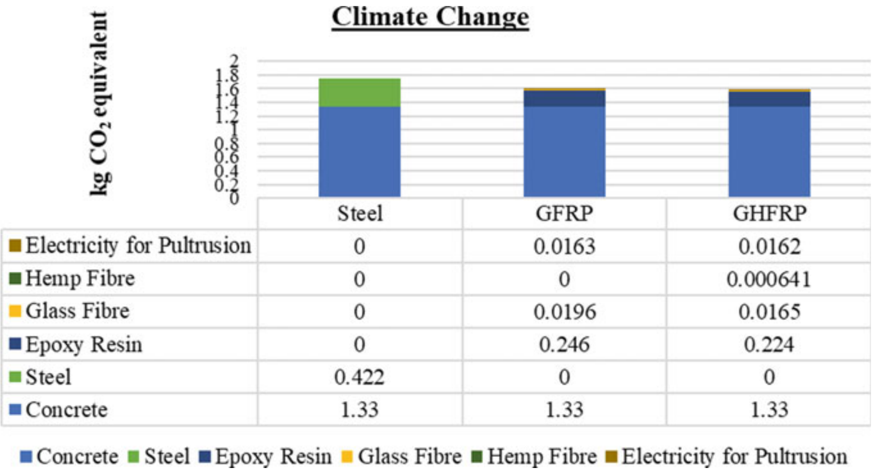
	Steel	GFRP	GHFRP
Electricity for Pultrusion	0	0.00467	0.00465
Hemp Fibre	0	0	0.000184
Glass Fibre	0	0.00673	0.00567
Epoxy Resin	0	0.0936	0.0852
Steel	0.115	0	0
Concrete	0.109	0.109	0.109

Fig. 1 Contribution of the constituents in RC beam systems on fossil depletion category

After substituting the steel rebar by GFRP, the value of fossil depletion in the new structure decreased, as expected. The contribution of concrete remained unchanged in both beam structures, meaning that the reduction of the 4.5% came entirely from benefit of replacing the steel rebar. Although having a comparably high energy demand as that in steel rebar in the production phase, the E-glass fiber gave a very low contribution to this impact category. This can be explained by the fact that only a very small amount of E-glass fiber material was required in the GFRP rebar to achieve the desired strength, due to its strong mechanical properties, particularly the tensile strength. In fact, the matrix phase, which was the epoxy resin, was regarded as the major contributor in GFRP to fossil depletion, based on Fig. 1. This petroleum-based polymer gave a share as high as 44% from the total value of fossil depletion, in which the value (0.0936 kg oil eq.) was comparable with that of steel rebar (0.115 kg oil eq.). A big concern arose here when it was found out that a small amount of epoxy resin (0.03 kg) can have an effect which was almost as significant as that by a big amount of steel rebar (0.33 kg). The main reason for this was because the major ingredient used in the production of epoxy resin was petroleum, which was actually the fossil fuel (Zaman et al. 2014). In other words, fossil fuel, in this case, was not only used to generate the energy required by the manufacturing processes involved but was also used as the raw material in the meantime.

3.2 Climate Change

Taking about the steel reinforced concrete beam, Fig. 2 illustrated that the impact caused by the concrete beams on climate change was, again, mainly contributed by concrete, which shared about 76% of the total value of this impact indicator, with



	Steel	GFRP	GHFRP
Electricity for Pultrusion	0	0.0163	0.0162
Hemp Fibre	0	0	0.000641
Glass Fibre	0	0.0196	0.0165
Epoxy Resin	0	0.246	0.224
Steel	0.422	0	0
Concrete	1.33	1.33	1.33

Fig. 2 Contribution of the constituents in RC beam systems on climate change category

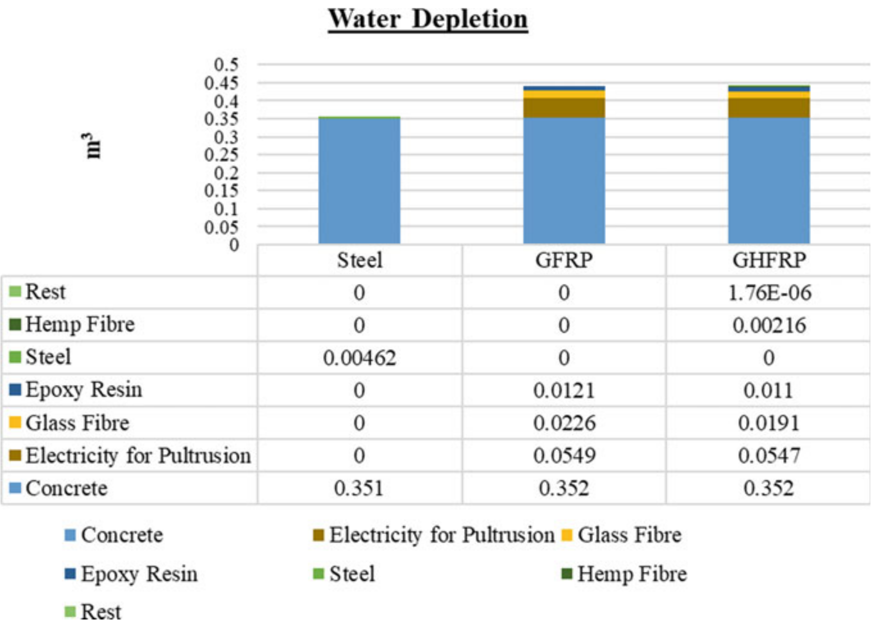
the remaining contributed by the steel rebar. This was easily explainable by the huge amount of the concrete material used (more than 97% of the total mass) in the beam structure itself that leaded to the higher contribution. In the material production wise, the manufacturing process of the ingredients of concrete, particularly the production of clinker used in the cement, was proven to have significant influence on the life cycle of concrete.

The manufacture of clinker involved the high consumption of greenhouse gases emitting materials as well as the burning of fossil fuels, and hence, the high environmental impact by the concrete material (Zabalza Bribián et al. 2011). In fact, a close relationship can be established between the climate change and the consumption of energy resources such as fossil fuels in the energy generation. To justify this, burning of fossil fuels will result in the emission of greenhouse gases to the air, especially the carbon dioxide, which was the major contributor of greenhouse gases in the atmosphere. Comparing Figs. 1 and 2, it was found out that the trend of the relative contribution of the individual constituents in the beam structure was comparable for both climate change and fossil depletion. Therefore, it can be deduced that the severity on climate change category corresponded to the energy requirement of the material production, meaning that the higher the energy required, the greater the negative effect on the climate change.

After replacing the steel rebar by GFRP, the contribution of the concrete was again found out to be similar as that in the case of steel reinforced concrete beam. With that said, the use of GFRP as the reinforcing component played a major role in the total decrease of 8%. This was explainable by the materials used in the steel rebar production. Different types of carbon-based combustion sources such as coke, coke breeze and coke oven gas were consumed heavily in the process, as presented in the LCA study by Burchart-Korol (2013). Also, as proven in the similar source, the processes such as sinter plant, blast furnace and hot rolling also emitted a huge amount of carbon dioxide. Focusing on the GFRP itself, most of the impact contributed by this composite, again, came from the epoxy resin. One of the reasons can be due to the higher relative mass of epoxy resin content (75% by mass) used in the GFRP as compared with that of glass fiber. From the perspective of material production process, La Rosa and Cicala (2015) argued that the fabrication of polymer matrix such as epoxy resin also required high energy content, especially the heat energy used for the melting of polymer. As mentioned earlier, high energy consumption can directly lead to higher amount of fossil fuels to be combusted for generating the enough energy and hence, more greenhouse gases were released in the process. Also, the production of the petroleum, the major feedstock used in the epoxy resin, also emitted a huge amount of greenhouse gases into the atmosphere, which justified the high contribution of this polymer matrix to climate change category.

3.3 Water Depletion

Based on the inventory data collected, both steel rebar and concrete required water in the manufacturing processes. However, in the steel reinforced concrete beam, the significance of the effect contributed by concrete alone to this impact category is obvious, as presented in Fig. 3. This can be first justified by the mass of the concrete used, as it took up approximately 97% of total mass in the beam structure. From the



	Steel	GFRP	GHFRP
Rest	0	0	1.76E-06
Hemp Fibre	0	0	0.00216
Steel	0.00462	0	0
Epoxy Resin	0	0.0121	0.011
Glass Fibre	0	0.0226	0.0191
Electricity for Pultrusion	0	0.0549	0.0547
Concrete	0.351	0.352	0.352

Fig. 3 Contribution of the constituents in RC beam systems on water depletion category

perspective of concrete production, water was also recognized as one of the major ingredients in concrete material and therefore, the higher the mass of the concrete used, the greater the amount of water was required for the production and hence, the bigger contribution from the environmental point of view.

Conversely, Fig. 3 showed that the total value of water depletion in the case of GFRP reinforced concrete beam was comparatively higher. On the same figure, it can be observed that the impact caused by GFRP reinforcing bar was significantly higher than of steel rebar. The additional value on the impact mostly came from the production of electricity required for pultrusion process as well as the fabrication of glass fiber. Scrutinizing on the individual constituents, the major contributor to this impact category was, again, the concrete, giving a share of around 80%, followed by pultrusion and glass fiber, with a share from the total value of about 12 and 5% respectively. In fact, the magnitude of effect contributed by concrete in this case was slightly higher than that in steel reinforced concrete beam. To explain this, FRP rebar was a stronger reinforcement generally as compared to the steel rebar, resulting in smaller reinforcement volume and greater amount of concrete used in the beam structure and hence, higher effect on the water depletion. Although, this increase of impact was insignificant due to the very small increase of the mass of concrete in our study, and therefore, it can be deduced that the effect by the water used for electricity production overrode that of the water used for concrete. Hereby, the focus here should be directed to the effect contributed by the production of electricity used for pultrusion and glass fiber fabrication. The power generation plant required the use of equipment such as heat exchangers, which leaded to the requirement of process cooling water for accomplishing the process. These waters can be extracted from various sources such as rivers, seas, groundwater and others (Zabalza Bribián et al. 2011). Therefore, high electricity requirement by the pultrusion process and glass fiber production resulted in higher demand of cooling water and hence, the higher water depletion value in this concrete beam. In some occasions, the cooling water might get evaporated after absorbing sufficiently high amount of heat, which further contributed some impacts onto the water footprint (Zabalza Bribián et al. 2011). Comparing the LCA inventories of steel rebars and E-glass fiber, the water usage was also much higher in the production phase of latter material, and hence, the higher impact (Thinkstep 2016).

3.4 Hybrid GHFRP RC Beam System

The variation of the constituent's composition in the hybrid GHFRP reinforced concrete beam before and after the inclusion of a small amount of natural hemp fiber has been reviewed in previous section. This change has shown noticeable benefits from the environmental point of view, and it was mostly attributable to the reduction of both E-glass fiber and epoxy resin contents. As compared with GFRP reinforced concrete beam, this concrete system scored better for climate change, fossil depletion and water depletion categories while it came short regarding natural land

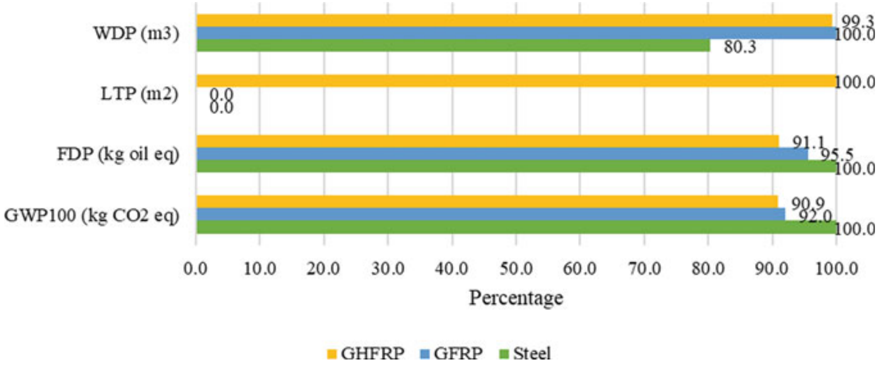


Fig. 4 Comparison of beam systems on four focused impact categories

transformation category. Again, the discussion here would be focusing on the impacts caused by the reinforcing component itself only. This was because the study has been simplified by intentionally making the tensile strength of both GFRP and GHFRP to be equal, which leaded to the unchanged mass of concrete used and hence, the similar environmental impacts by concrete in both systems.

From the perspective of climate change and fossil depletion, the addition of hemp fiber gives a further reduction of approximately 1.1 and 4.5% respectively, as presented in Fig. 4. The contribution of the hemp fiber in here was negligible due to the very little amount of the hemp fiber used in the concrete beam and because of the natural fiber had extremely low dependence on non-renewable energy resources as well as insignificant greenhouse gases emission (La Rosa et al. 2013b). Scrutinizing on the greenhouse gases emission itself, La Rosa et al. (2013b) and La Rosa and Cicala (2015) also mentioned that the natural fibers are generally carbon neutral as the plants, where the fibers are derived from, absorb carbon dioxide during the vegetative cycle and therefore, they give insignificant or no impact on the climate change. However, the positive impacts achieved in this beam system were mainly due to the reduction in the amount of both E-glass fiber and epoxy resin used in the hybrid GHFRP. For instance, by adding merely 4.03 g of hemp fiber, the mass of E-glass fiber dropped by around 16% and similar case is also observed in the mass of epoxy resin, which gave a reduction of around 9%. Considering that the fabrication of E-glass fiber was a very high-energy-demand process, replacing some of the amounts of this material by low energy natural fiber like hemp fiber directly lowered the energy requirement during the production phase. However, the combustion of fossil fuel for power generation was significantly lessened, which ultimately leaded to lower greenhouse gases emission and fossil fuel consumption, and therefore, lower impacts to both climate change and fossil depletion. Similarly, from the perspective of epoxy resin, lower material content meant that the energy requirement of the manufacturing process was decreased directly. Also, considering that the fossil fuel was one of the major feedstocks of this polymer matrix, requiring lower amount of epoxy resin guaranteed lower consumption of this depleting resources and hence, the decreased effects on both impact categories can again be justified. These findings were comparable with those presented in the LCA case study conducted by Corbière-Nicollier et al. (2001).

As for the water depletion category, the use of GHFRP also gave a slight decrease of 0.7% as compared with that of GFRP. Again, this was mostly attributable to the reduction of glass fiber and epoxy resin contents as well as the reduction of mass in the reinforcing bar itself as it can be observed in Fig. 3 that the values contributed by all the constituents on this impact category decreased. Lower amount of materials used and reduced mass of reinforcing bar leaded to lower electricity requirement for the fabrication processes of these materials as well as the pultrusion process and therefore, less process water was required in this case. In the similar figure, it can be observed that only the production of electricity used in the hemp fiber processing contributed to this impact category. In other words, the cultivation stage of hemp fiber plants did not have any effect on water depletion potential. La Rosa et al. (2013b) explained that, unlike other natural fibers, water irrigation is not required for the cultivation of hemp fiber as the natural rainfall during the vegetative stage is enough for the growth of hemp plants.

Apart from decreasing the amount of the constituents used in the hybrid GHFRP, the inclusion of hemp fiber also helped in lowering the mass of the reinforcement itself in overall, as mentioned earlier. As an example, the mass of the reinforcing bar lowered slightly by approximately 0.65% when GHFRP was used to replace the GFRP. The finding was justifiable based on the argument presented by the La Rosa et al. (2013b) in their LCA case study on weight saving benefit using natural fibers. The benefit of this mass reduction can be seen in the pultrusion process of FRP rebars. It can be deduced that the lower the FRP mass, the lower the electricity required to manufacture the FRP reinforcing bars since the electricity requirement of this process was dependable on the mass of the FRP, which consumed 3.1 MJ per 1 kg of FRP rebar, as presented by Song et al. (2009). Therefore, the use of GHFRP in the concrete beam has demonstrated its environmental advantages in three of the impact categories mentioned earlier.

On the contrary, the use of GHFRP rebar in the concrete beam has, in turn, brought about negative impact on the natural land transformation category. To produce 4.03 g of hemp fiber used for the concrete system in Scenario 1, 2.81×10^{-6} m^2 of land occupation was required and the value was entirely contributed by the hemp fiber itself. This phenomenon was quite predictable as the forest area or natural land had to be transformed into arable agricultural land for the cultivation and cropping of natural hemp fiber. Concrete beams reinforced by both steel and GFRP rebars did not require any land occupation, meaning that there was zero impact from the production of all steel, concrete, glass fiber and epoxy resin on this impact category. In fact, this was justifiable because the natural land required by the fabrication process of these materials, as compared with that of hemp fiber, was very little and insignificant. The similar finding was also presented by La Rosa et al. (2014) in their LCA case study that producing 1 kg of hemp mat required a land area as much as 1.54 m^2 while only 0.0692 m^2 of land was needed for producing the similar mass of glass fiber, which was only 4.5% of that of hemp mat.

4 Conclusion

Over the last decade, Use of FRP composites in the construction industry have been gaining attention due to conventional constructive materials such as steel and cement contribute to high environmental impacts. Thus, a significant research has been done to investigate possibility of replacing existing construction materials with FRP composites. The concrete beam reinforced by hybrid GHFRP was comparatively more eco-efficient than that of GFRP and steel rebar. From the results, it was notable that the major contribution to the impact, in all cases, was due to concrete. In FRP case, the governing aspect on the environmental impacts was the amount of epoxy resin used in the structure. The valuable ecological advantages received from the inclusion of hemp fiber that reduced the relative amounts of other higher-impact-constituents and the mass of the reinforcing bar itself have also been observed. By examining the effects of the composition variation, it came to a conclusion that the environmental standing of this concrete beam system improved when the amount of epoxy resin and hemp fiber are reduced and increased respectively The existing LCA case studies on different materials were also critically reviewed for better understanding and based on which, it was discovered that LCAs conducted on both traditional and newly-invented materials such as composites in various industries have gradually emerged in recent years. Although, the popularity of LCA particularly in the use of composites in construction industry has been very low, due to the inherent complexities associated with the building itself. Knowing the importance of proper material selection and the viability of using FRP in several building applications, a huge research gap has been created for future researchers in conducting LCA on replacing the conventional materials with this new material, to understand fundamentally how this change or revolution can affect the environment positively and negatively.

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Dielectric Properties of Acacia Wood Bio-composites

M. K. B. Bakri, P. L. Nyuk Khui, Md Rezaur Rahman, S. Hamdan, E. Jayamani and A. Kakar

Abstract In this chapter, pure PLA, PHA, PLA/PHA (1:1 ratio of PLA/PHA), and NCHB-PLA/PHA (1 wt% of NCHB; 1:1 ratio of PLA/PHA) with addition of different fiber loadings (i.e. 5 wt%, 10 wt%, 15 wt% and 20 wt%) are fabricated. It is found that the addition of Acacia wood (AW), caused an inrease in the dielectric constant, dissipation factor and loss factor. The highest optimum value was obtained at 20 wt% fiber loadings. Addition of AW fiber caused an increase in the dielectric constant, dissipation factor and loss factor. Chemical modification on the AW fiber were also done and resulted, which resulted in the increase of hydrophobicity of the modified AW fiber. Polymer blend created better interlocking, reduce the formation of bubble/void, which create low dielectrical properties than PLA and PHA itself. Addition of NCHB stabilizes the dielectric constant, dissipation factor and loss factor, which led to a smoother data, when compared to other results.

Keywords Dielectric properties · Dielectric constant · Dissipation factor · Loss factor

1 Introduction

A material's dielectric constant (ε′) is the measure of its polarization and the charge stored as an external electric field was applied parallel to the plates that act as a capacitor (Mehta and Parsania 2006; Parida et al. 2015). Alternatively, Sreekumar et al. (2012) defined it as a ratio of the capacitance of the condenser in real condition

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M. K. B. Bakri · P. L. Nyuk Khui · Md R. Rahman (✉) · S. Hamdan
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

E. Jayamani
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

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over condenser under vacuum. A higher value of dielectric constant showed a greater degree of polarization.

In heterogeneous materials, interfacial polarization occurred, which is due to the fiber and matrix differences in conductivities (Parida et al. 2015). The interfacial polarization effect can be identified by its high dielectric constant at low frequency. There is also a possibility that at low frequency regions, the total orientation of polar molecules is high due to the orientation of polarization. On the other hand, at higher frequency, electrical dipole orientation restricts due to smaller time interval (Parida et al. 2015). Thus, the presence of fillers and polar group and externally applied electric field frequency influenced the dielectric constant (Chand and Jain 2005; Joseph and Thomas 2008).

The electric loss is known when the amount of energy dissipated when a voltage is applied in the circuit as the materials being insulated. Represented by the loss tangent, the energy dissipation was also known as dissipation factor (tan δ). It is calculated by the ratio of dissipation of electric power in the materials to the net power in the circuit. Tan δ measurement of an insulating material is important to understand the amount of electrical energy that is being converted into heat. Most of the heat accelerated its deterioration and increased the insulator temperature. Loss factor ($\acute{\varepsilon}$) is the average power factor over a given period of time. It also represented the distribution and transmission loss (Sreekumar et al. 2012). The polymer viscoelastic nature created responses in both mechanical and electrical stimuli of the material. The loss factor ($\acute{\varepsilon}$) two prime factors are the ionic conductance and dipole polarization (Chand and Jain 2005; Jacob et al. 2006).

2 Experimental

2.1 Materials

The Acacia wood (AW) from *Acacia mangium* species was provided by Sarawak Forestry Corporation Sdn. Bhd. (SFCsb), Kuching, Sarawak, Malaysia. The AW fibers lengths are less than 3.5 mm. It has a density range of 420–483 kg/m^3 (in green soaked volume) and 500–600 kg/m^3 (in dry condition) (Logan and Balodis 1982; Peh et al. 1982; Peh and Khoo 1984; Razali and Kuo 1983; Wang et al. 1989). Naturally grown AW has a specific gravity (SG) of 0.56 kg/m^3, while the re-plantation grown AW has a lower SG of 0.40–0.45 kg/m^3 (National Research Council 1983). However, in Borneo, the SG can specifically be obtained around 0.43–0.47 kg/m^3 for 6-years old AW (Sining 1989).

For naturalization modification technique, two types of chemicals were used; acid and alkaline. The pure acetic acid (CH_3COOH) solution was obtained from Fisher Scientific (UK), Ltd., known by the product name “Acetic Acid, ACROS Organics$^{\text{TM}}$”. Powdered sodium bicarbonate ($NaHCO_3$) was obtained from Fisher Scientific (UK), Ltd., known by the product name “Sodium Bicarbonate, ACROS Organics$^{\text{TM}}$”.

Polyhydroxyalkanoates (PHA) was obtained from Jinan Pu Dong Import and Export Trading (China) Co. Ltd. with chemical abstracts service (CAS) number "718630-59-2". It had a density of 1.2–1.4 kg/m^3 and contained 90% bio-based carbon according to ASTM D6866-16 (2016) standard. Pure grade powdered PLA was obtained from Shenzhen Esun Industrial (China) Co. Ltd. with product code "Al-1001". It had 100% bio-based carbon according to the ASTM D6400-12 (2012) standards. It had a density of 1.25 kg/m^3 and melt flow index (MFI) of 10–12 g/10 min.

2.2 Fiber Preparation

For unmodified fiber, the AW fibers were washed and rinsed with distilled water. Later, it was dried in an open ventilated oven at 80 °C for 24 h. The ventilated oven model "ECOCELL EC55" equipped by MMM Medcenter Einrichtungen (Germany) GmbH was used in this process.

This process involved the use of two different man-made bio-based chemicals. The AW fibers were soaked for 24 h inside 1.5 L of 5 wt% of acetic acid solution (CH_3COOH) mixed with distilled water. After that, 90gm of sodium bicarbonate ($NaHCO_3$) was poured into the soaked AW for 24 h. The reactions of all possible chemicals are as below:

$$\mathrm{Na_2CO_3 + 2CH_3COOH \rightarrow 2CH_3COONa + H_2O + CO_2} \tag{1}$$

$$\mathrm{Na_2HCO_3 + CH_3COOH \rightarrow NaCH_3COO + H_2O + CO_2} \tag{2}$$

$$\mathrm{Wood - OH + CH_3COONa \rightarrow Wood - O - NaH + CH_3COO} \tag{3}$$

The short AW fibers were filtered and rinsed using distilled water to neutralized remaining chemicals attached to the AW fibers. Lastly, it was dried in an open ventilated oven at 80 °C for 24 h. All the materials obtained were concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.3 Fabrication of the Bio-composites

Pure PLA, PHA, PLA/PHA (1:1 ratio of PLA/PHA), and NCHB-PLA/PHA (1 wt% of NCHB; 1:1 ratio of PLA/PHA) with addition of different fiber loadings (i.e. 5, 10, 15 and 20 wt%) are fabricated. The mixtures of the ingredients were done using a drying mixer machine. It was mix blend for about 5 min. The molds used for the bio-composites fabrication, were designed according to ASTM D150-11 (2011) for dielectric properties. The molds were cleaned and waxed before they were filled in with the mixture of composite materials (i.e. fiber, polymer and nano-enhancement).

Waxes were applied on the molds. The releasing wax contained carnauba that prevents the sample sticks and stuck. Aluminum foils were placed in between the plates, which made the samples easier to be removed from the molds and reduced the formation of bubbles in the samples. The mixtures of composite materials were placed in the mold frame according to the fiber ratio. The mold was compressed using a hot press hydraulic machine model 'LS-22071' equipped by Lotus Scientific (Malaysia) Sdn. Bhd. under pressure of 1000 psi (6.89 MPa). It was then placed in the hot press machine and heated at 150 °C for 6 min. The samples were then quenched in water at room temperature, according to ASTM E41-92 (2010). The samples were then dried using a hair dryer and were kept concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.4 Methods

2.4.1 Dielectric Test

The dielectric tests were done according to ASTM D150-11 (2011) standards using a dielectric test fixture. The machine was supplied by Agilent Technologies Microwave Products (Malaysia) Sdn. Bhd. model 'Agilent E4980A precision LCR meter'. The dielectric constant measurement was carried out using dielectric test fixture model '16451B'. The LCR meter can measure up to 2 MHz. The disc-shaped samples had a diameter of about 50 mm and a thickness of about 5 mm. The samples were analyzed using a contacting electrode method, which uses a rigid metal electrode. The measurements were done from 1 kHz to 2 MHz. Each of the samples was tested 5 times at a given frequency and then the average value was recorded by the HP impedance analyzer. The dielectric constant was calculated as below (Markiewcz et al. 2009):

$$\varepsilon' = \frac{C_p d}{\varepsilon_0 A} \tag{4}$$

where ε' is the dielectric constant, C_p is the sample capacitance, d is the thickness, ε_0 is the vacuum permittivity 8.854×10^{-12} F/m and A is the circular area. The dissipation factor was calculated as below:

$$\theta = \tan \delta \tag{5}$$

where θ is the dissipation factor, δ is the loss angle. The loss factor was calculated as below:

$$\varepsilon'' = \varepsilon' \theta \tag{6}$$

where, ε'' is the loss factor.

3 Results and Discussion

3.1 Effects of Fiber Loading on PLA and PHA Bio-composites Dielectrical Properties

3.1.1 Dielectric Constant

Figures 1 and 2 shows the dielectric constant graph for U-AW-PLA, M-AW-PLA, U-AW-PHA, and M-AW-PHA bio-composites at different fiber loadings. It was observed that the dielectric constant increases, as the fiber content increase. The lowest dielectric constant was exhibited by pure PLA and PHA. This was due to the absence of fiber, which contributes to permanent dipoles, since matrix was composed by carbon and hydrogen only. PHA had higher dielectric constant than that of PLA. This may be due to the higher density of PHA, which implies more closely packed molecules and lesser micro-voids.

Highest dielectric constant was at 20 wt% fiber loadings. It is known that the dielectric constant depends on the orientation and interface of the fiber, and electronic and atomic polarization in the bio-composites. According to Haseena et al. (2007), the interfacial polarization occurred due to the differences in conductivities or polarizations of the matrix

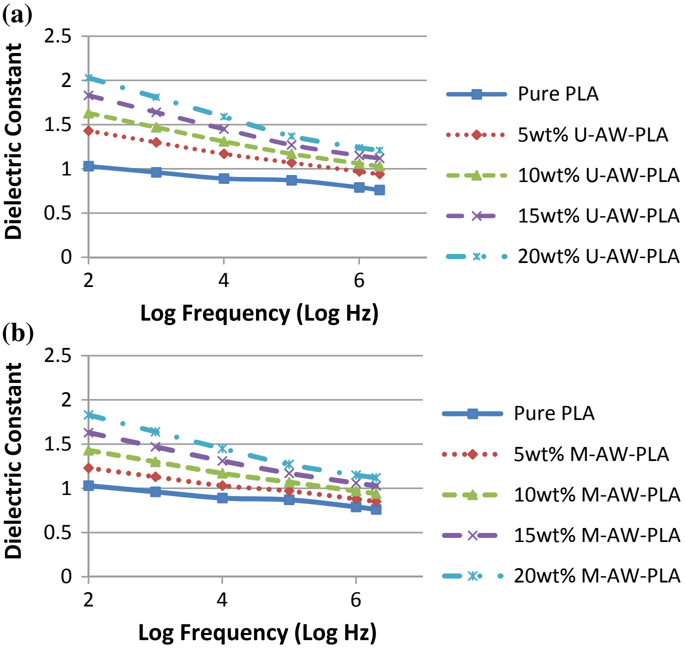

Fig. 1 Dielectric constant graph of **a** U-AW-PLA and **b** M-AW-PLA

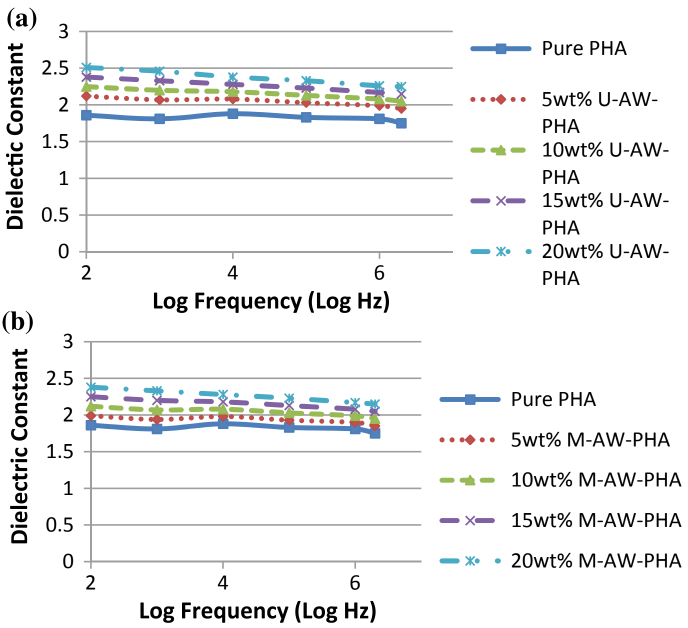

Fig. 2 Dielectric constant graph of **a** U-AW-PHA and **b** M-AW-PHA

and fibers. Thus, composite dielectric constant properties are dependent on the interfacial and orientation of the fibers. With the increase in fiber loading, the significant increment in dielectric constant was due to the increment in orientation and interfacial polarization resulting from the presence of polar groups of cellulose in AW fiber (Haseena et al. 2007). Increase in the conductivity of the polymer materials may also be due to the hydrophilic nature of the AW fiber, which has the tendency to absorbed moisture content from the air (Singha et al. 2013).

3.1.2 Dissipation Factor

Figures 3 and 4 shows the dissipation factor graph for U-AW-PLA, M-AW-PLA, U-AW-PHA, and M-AW-PHA bio-composites at different fiber loadings. The dissipation factor was found to decrease with the increase of frequencies. It was also observed that the dissipation factor increases with the increase in the fiber content. The lowest dissipation factor was exhibited by pure PLA and PHA. This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. PLA had higher dissipation factor compared with PHA, which was due to the amorphous region which allowed and enhanced the current flow through the region. The highest dissipation factor was at 20 wt% fiber loadings. According to Sreekumar et al. (2012), the increment in the orientation polarization was due the increase number of polar groups.

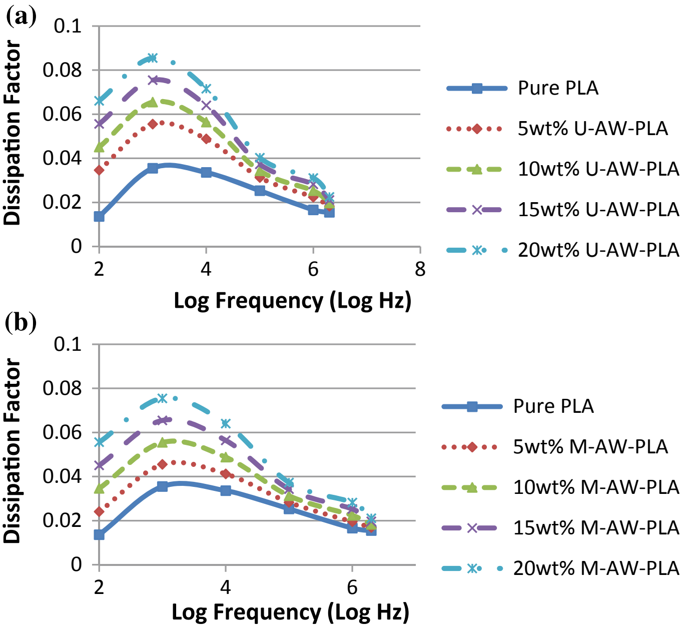

Fig. 3 Dissipation factor constant graph of **a** U-AW-PLA and **b** M-AW-PLA

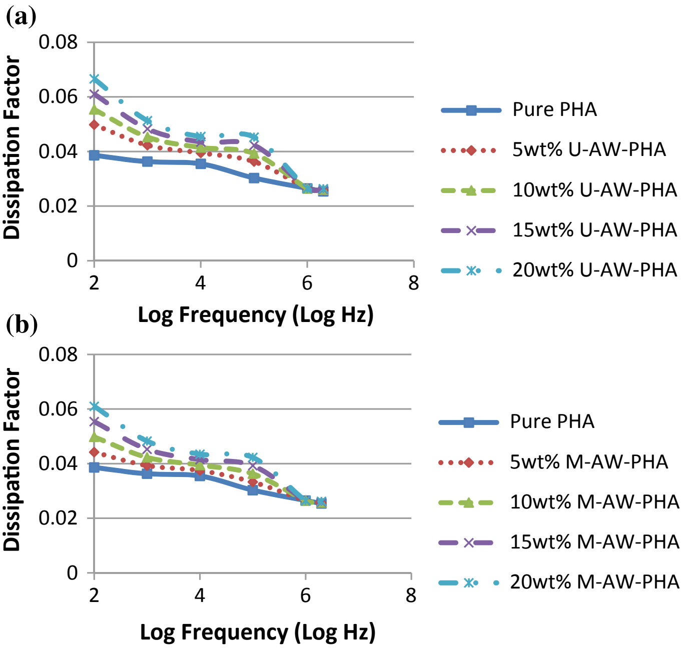

Fig. 4 Dissipation factor graph of **a** U-AW-PHA and **b** M-AW-PHA

3.1.3 Loss Factor

Figures 5 and 6 show the loss factor graph for U-AW-PLA, M-AW-PLA, U-AW-PHA, and M-AW-PHA bio-composites at different fiber loadings. It was observed that the loss factor increased as the fiber content increased. The lowest loss factor was exhibited by pure PLA and PHA. This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. PLA is also noticed to have higher loss factor compared with PHA, which was due to the amorphous region which allowed and enhanced the flow current through the region. The highest loss factor was at 20 wt% fiber loadings. According to Sreekumar et al. (2012), the increment in the orientation polarization was due the increase number of polar groups. However, the exact explanation is difficult to explain as it depends upon other behaviors such as fiber orientation, in which the fibers were inserted in the composites in such random orientation and moisture may also be trapped and contributed to such behaviors (Singha et al. 2013).

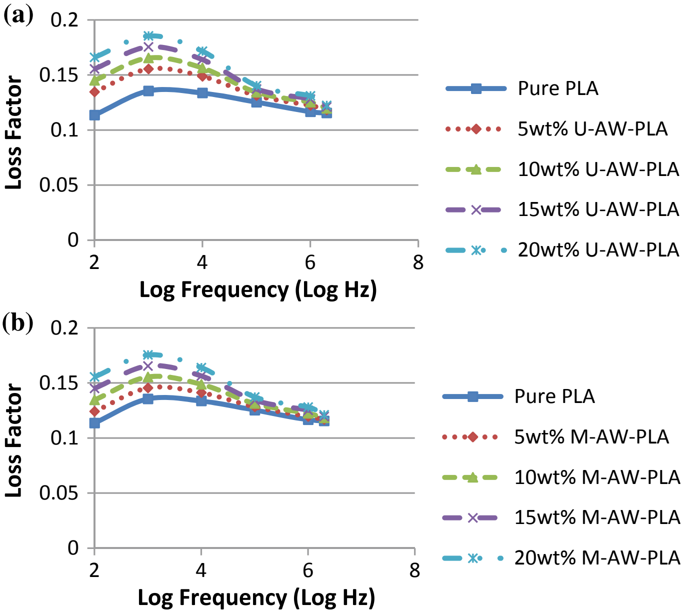

Fig. 5 Loss factor constant graph of **a** U-AW-PLA and **b** M-AW-PLA

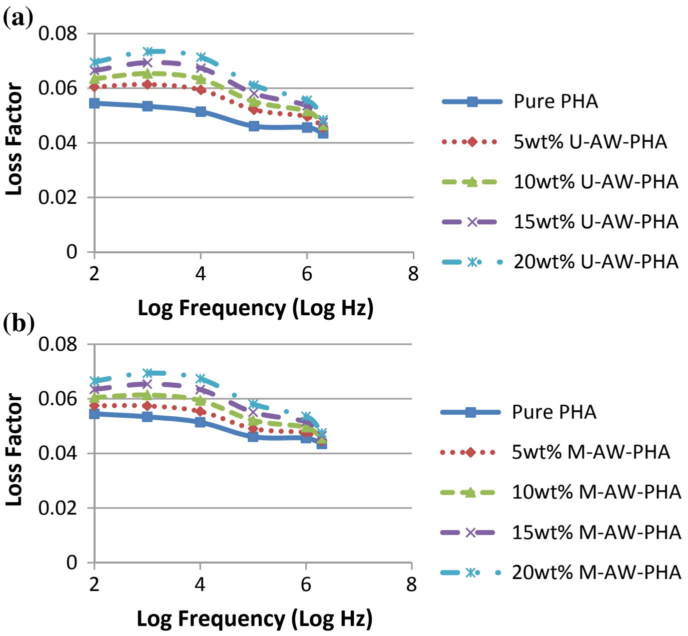

Fig. 6 Loss factor graph of **a** U-AW-PHA and **b** M-AW-PHA

3.2 Effects of Polymer Blend (PLA/PHA) on Bio-composites Dielectrical Properties

3.2.1 Dielectric Constant

Figure 7 shows the dielectric constant graph for U-AW-PLA/PHA, and M-AW-PLA/PHA bio-composites at different fiber loadings. It was observed that the dielectric constant increased as the fiber content increased. With the increase in the fiber loading, the significant increment in dielectric constant was due to the increment in orientation and interfacial polarization resulting from the presence of polar groups of cellulose in AW fiber (Haseena et al. 2007). The lowest dielectric constant was exhibited by pure polymer blend (PLA/PHA). This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed only carbon and hydrogen. Compared with PLA and PHA, the polymer blend had low stable dielectric constant, which was due to the interlocking between PLA and PHA. The structural properties of semi-crystalline/semi-amorphous structure caused sharing of the dipoles, which led to a more stable dielectric constant (Thakur et al. 2017). Highest dielectric constant is at 20 wt% fiber loadings. It is known that the dielectric constant depends on the orientation and interface of the fiber, and electronic and atomic polarization in the bio-composites.

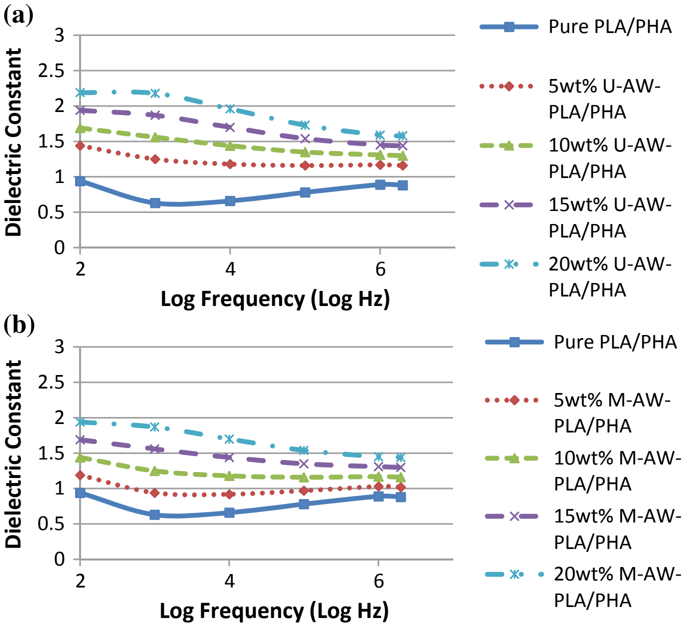

Fig. 7 Dielectric constant graph of **a** U-AW-PLA/PHA and **b** M-AW-PLA/PHA

3.2.2 Dissipation Factor

Figure 8 shows the dissipation factor graph for U-AW-PLA/PHA, and M-AW-PHA/PHA bio-composites at different fiber loadings. It was observed that the dissipation factor increased as the fiber content increased. The lowest dissipation factor was exhibited by pure polymer blend (PLA/PHA). This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. Compared with PLA and PHA, the polymer blend had lower stable dissipation factor, which was due to the interlocking between PLA and PHA. Highest dissipation factor is at 20 wt% fiber loadings. According to Sreekumar et al. (2012), the increment in the orientation polarization was due to the increase number of polar groups.

3.2.3 Loss Factor

Figure 9 shows the loss factor graph for U-AW-PLA/PHA, and M-AW-PLA/PHA bio-composites at different fiber loadings. It was observed that the loss factor increased as the fiber content increased. The lowest loss factor was exhibited by pure polymer blend (PLA/PHA). This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. Compared with PLA and PHA, it is noticed that the polymer blend had higher stable

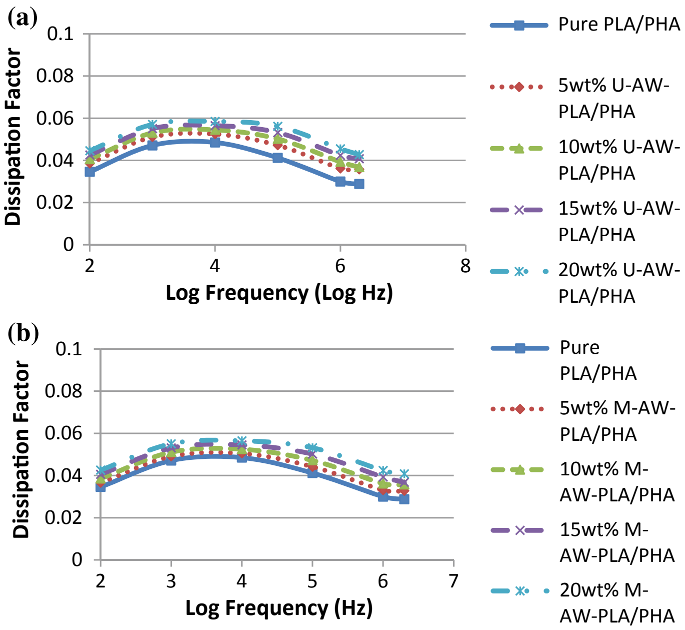

Fig. 8 Dissipation factor constant graph of **a** U-AW-PLA/PHA and **b** M-AW-PLA/PHA

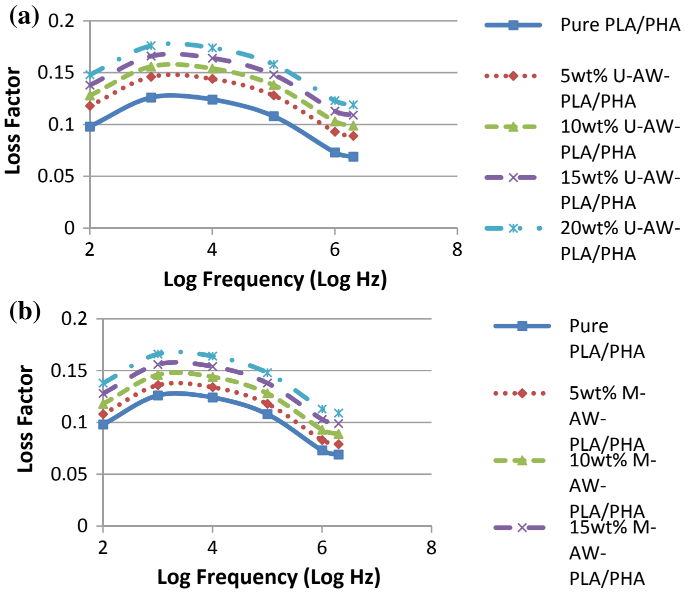

Fig. 9 Loss factor constant graph of **a** U-AW-PLA/PHA and **b** M-AW-PLA/PHA

loss factor constant. Highest loss factor is at 20 wt% fiber loadings. As the fiber content increases, the heterogeneity of the composite increases, thereby increase its polarizations (Sreekumar et al. 2012).

3.3 *Effects of Nano-enhancement on Polymer Blend (PLA/PHA) Bio-composites Dielectrical Properties*

3.3.1 Dielectric Constant

Figure 10 shows the dielectric constant graph for U-AW-NCHB-PLA/PHA, and M-AW-NCHB-PHA/PHA bio-composites at different fiber loadings. It was observed that the dielectric constant increases with an increase in the fiber content. With the increase in the fiber loading, the significant increment in dielectric constant was due to the increment in orientation and interfacial polarization resulting from the presence of polar groups of cellulose in AW fiber (Haseena et al. 2007). Addition of NCHB in the polymer blend bio-composites leads to a more stable dielectric constant (with slight increase as frequency increase) compared with polymer blend

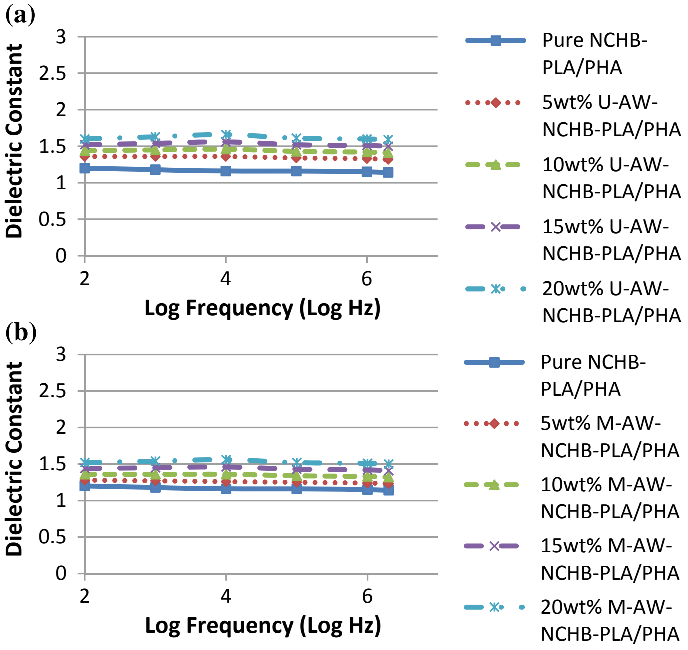

Fig. 10 Dielectric constant graph of **a** U-AW-NCHB-PLA/PHA and **b** M-AW-NCHB-PLA/PHA

bio-composites itself. Highest dielectric constant is at 20 wt% fiber loadings. As the fiber content increases, the heterogeneity of the composite increases, thereby increase its polarizations (Sreekumar et al. 2012).

3.3.2 Dissipation Factor

Figure 11 shows the dissipation factor graph for U-AW-NCHB-PLA/PHA, and M-AW-NCHB-PHA/PHA bio-composites at different fiber loadings. It was observed that the dissipation factor increases with an increase in fiber content. The lowest dissipation factor was exhibited by pure NCHB polymer blend (PLA/PHA). This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. Addition of NCHB in the polymer blend bio-composites caused more stable dissipation factor compared with polymer blend bio-composites itself. Highest dissipation factor is at 20 wt% fiber loadings. As the fiber content increased, the heterogeneity of the composite increased, thereby increase its polarizations (Sreekumar et al. 2012).

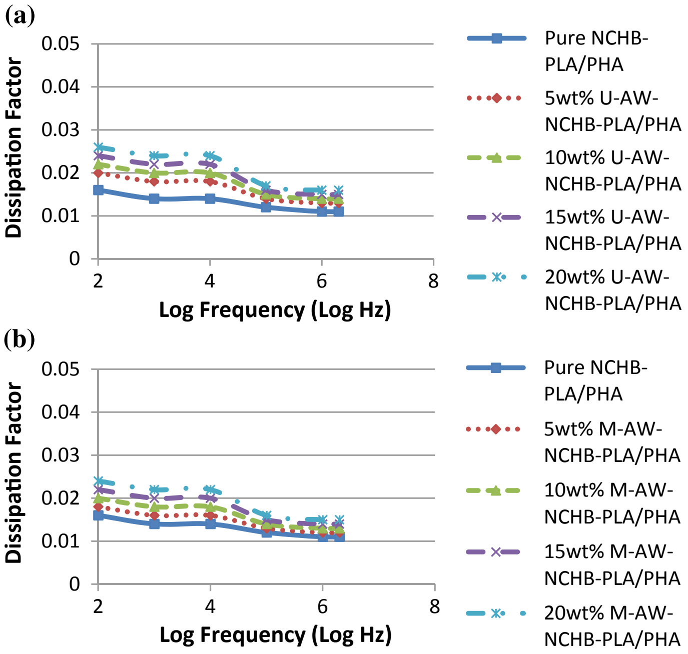

Fig. 11 Dissipation factor constant graph of **a** U-AW-NCHB-PLA/PHA and **b** M-AW-NCHB-PLA/PHA

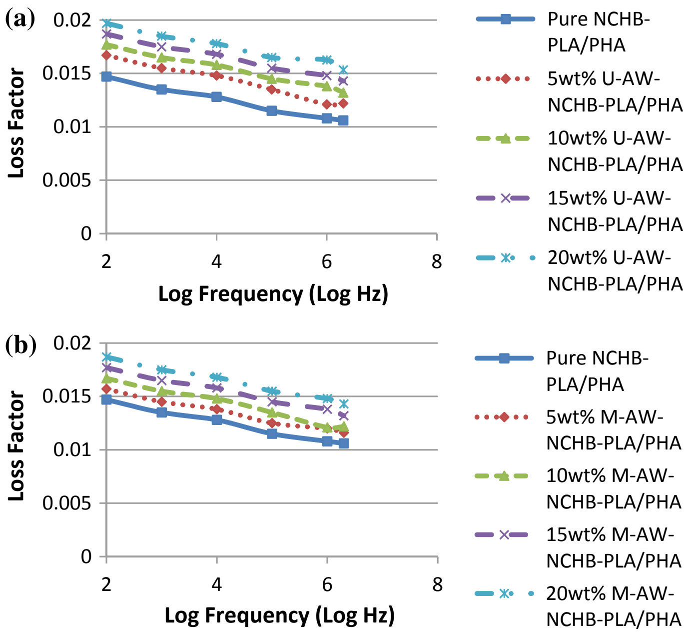

Fig. 12 Loss factor constant graph of **a** U-AW-NCHB-PLA/PHA and **b** M-AW-NCHB-PLA/PHA

3.3.3 Loss Factor

Figure 12 shows the loss factor graph for U-AW-NCHB-PLA/PHA, and M-AW-NCHB-PHA/PHA bio-composites at different fiber loadings. It was observed that the loss factor increases with an increase in fiber content. The lowest loss factor was exhibited by pure NCHB polymer blend (PLA/PHA). This was due to the absence of fiber, which contributed to permanent dipoles, since matrix was composed by carbon and hydrogen only. Addition of NCHB in the polymer blend bio-composites causes more stable loss factor compared with polymer blend bio-composites itself. Highest loss factor is at 20 wt% fiber loadings. As the fiber content increases, the heterogeneity of the composite increases, thereby increase its polarizations (Sreekumar et al. 2012).

3.4 Effects of Chemical Treatment on AW Fiber as Reinforcement in PLA, PHA, Polymer Blend (PLA/PHA) and NCHB Polymer Blend Bio-composites Dielectrical Properties

It was observed that the chemically modified AW fiber had lower dielectric constant, dissipation factors and loss factor. This was due to the decrease in orientation polarization as resulted of increased hydrophobicity of the modified fibers. Chemical modification causes modified fiber −OH group to be reactive due to removal of hydrogen bond compared with unmodified fiber which is relatively unreactive, since hydrogen bonds is formed (Sreekumar et al. 2012). The hydrophilicity of the fibers gets reduced by treatment, in which decreases the orientation of polarization (Sreekumar et al. 2012). Another reason was due to removal of lignin, hemicellulose and cellulose, which reduced the size of the fiber, which cause reduction in polarization of the bio-composites (Sreekumar et al. 2012).

4 Conclusion

In this chapter, addition of AW fiber caused an increase in the dielectric constant, dissipation factor and loss factor. This was due to increase in the orientation and interface of the fiber, and electronic and atomic polarization in the bio-composites. Chemical modification on the AW fiber caused a decrease in the orientation polarization, which resulted in the increase of hydrophobicity of the modified AW fiber. Polymer blend created better interlocking, reduce the formation of bubble/void, which create low dielectrical properties than PLA and PHA itself. Addition of NCHB stabilizes the dielectric constant, dissipation factor and loss factor, which led to a smoother data, when compared to other results.

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Effect of Nano-enhancement on Acacia Wood Bio-composites

P. L. Nyuk Khui, Md Rezaur Rahman, S. Hamdan, M. K. B. Bakri, E. Jayamani and A. Kakar

Abstract This chapter explain about nano-enhancement on acacia wood bio-composites. The tensile, flexural and impact tests were describe in this study. The optimum tensile strength was achieved at 10 wt% weight percentage for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. The optimal flexural strength for AW-NCHB-PLA/PHA bio-composites was achieved at 10 wt% weight percentage for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. The optimal impact strength for AW-NCHB-PLA/PHA bio-composites was achieved at 10 wt% fiber loadings for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. Nano-clay enhancement increased the viscosity of the polymer blend, hence increases the threshold mechanical strength of the bio-composites. A strong adhesion was created between fiber and polymer, due to the incorporation of nano-clay, which reduces the agglomeration and formation of void during fabrication. It was observed that the U-AW and NCHB-PLA/PHA has a smooth surface structure and could be considered as semi-brittle/semi-ductile.

Keywords Nano-enhancement · Mechanical strength · Infrared spectrum · Thermal properties

1 Introduction

Due to the fiber structure, chemical modifications, etc., wood and other natural fiber reinforced polymer composites have gained a lot of popularity (Bledzki and Faruk

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P. L. Nyuk Khui · Md R. Rahman (✉) · S. Hamdan · M. K. B. Bakri
Faculty of Engineering, Universiti Malaysia Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

E. Jayamani
Faculty of Engineering, Computing and Science, Swinburne University of Technology Sarawak Campus, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

A. Kakar
Department of Materials Science and Engineering, University of Houston, 4800 Calhoun Road, Houston, TX 77204, USA

Md R. Rahman (ed.), *Acacia Wood Bio-composites*, Engineering Materials,
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2003). From the high demand of applications and being price driven, wood fibers was utilized as a reinforcement material in polymer bio-composites (Chuai et al. 2001; Eichhorn et al. 2001; Felix and Gatenholm 1991; Ismail and Mega 2001; Joseph et al. 1996; Maldas and Kokta 1990; Rana and Jayachandran 2000; Van de Velde and Kiekens 2001). Woods could be classified into hardwood and softwood. Examples of the types of hardwood are; Acacia (Acacia), Alder (Alnus), and Birch (Betula). For the types of softwood there are; Fir (Abies), Cedar (Cedrus), and Pine (Pinus).

Acacia mangium (AM) or *Acacia auriculiformis* (AA) are Leguminous tree species of the sub-family Mimosoideae, which could be classified as a type of hardwood. The Leguminous tree species are native and originally discovered in North Queensland Australia, Western Province of Papua New Guinea and the Moluccas East/Islands of Indonesia (Doran and Turnbull 1997). AM are found to be in the lowland areas and are important, as AM are usually used as a multipurpose tropical tree. The AM species was introduced as a firebreak barrier in Sabah in 1967.

Before the year of 2020 (PERKASA 2009), one million hectares of land in Sarawak were reserved for forest replantation and commercialized program. This decision was made in efforts to preserve and conserve the natural forest, it is also to meet current and future raw material demands in the timber industry.

These species have the potential of utilization for numerous applications within the timber industry, this is due to the species fast growth and adaptability towards the local climate conditions. During rehabilitation and reforestation, Acacia wood (AW) species could be utilized for restoration of soil conditions (Inagaki and Titin 2009; Yamashita et al. 2008; Yang et al. 2009). AW could be utilized for numerous timbers and non-timber products or applications. Examples of AW products or applications ranges from; furniture manufacturing, plywood, particleboard, fence posts, timbers, charcoal, pulp, adhesive, ornaments and architectural landscape purposes. Apart from AW timber, the leaves could be used as fodder for livestock and agricultural fertilizers. AW do not have a distinctive odor nor taste, the grain texture is fine and uniform, and interlocked with a medium lustre.

Clay is a mineral, which could be categorized into groups of silicates which are layered in structure. Most types of layered silicates are cheap and abundantly available and could be either natural or synthetic. Most layered silicates utilize for numerous applications are Nano-clay, where the layered silicates consisted of regular stack of aluminosilicate layers with a high aspect ratio and surface area. Typically, Nano-clay particles are extracted as according to soil fraction, with less than 2 μm in size with 1 nm thickness (Mittal 2009).

Nano-clay reinforced polymer composites has gain recognition due to its significant improved characteristics in terms of physical (i.e. permeability and shrinkage), thermal (i.e. decomposition, flammability, thermal stability and coefficient of thermal expansion, etc.) dielectrical (i.e. dielectric constant, loss factor, dissipation factor, etc.), morphological (i.e. optical, etc.) and mechanical (i.e. strength, modulus, etc.) properties (Lu and Mai 2005; Ma et al. 2003; Ray and Okamoto 2003; Yoonessi et al. 2004). According to Jiang et al. (2007), it was report that PLA could be toughened and reinforced simultaneously with a low concentration of MMT (<2.5 wt%). A similar study according to Chen et al. (2005) also reported MMT epoxy utilization in PLA/PBS/MMT ternary blend by melt blending showed similar modulus to neat PLA (1990 MPa versus 2215 MPa) and significantly increased the elongation at break (118.1% versus 6.9%).

2 Experimental

2.1 Materials

Acacia wood (AW) from Acacia mangium species was provided by Sarawak Forestry Corporation Sdn. Bhd. (SFCsb), Kuching, Sarawak, Malaysia. The length of the AW fibers is less than 3.5 mm and the density ranges from 420–483 kg/m^3 (in green soaked volume) and 500–600 kg/m^3 (in dry condition) (Logan and Balodis 1982; Peh et al. 1982; Peh and Khoo 1984; Razali and Kuo 1983; Wang et al. 1989). Naturally grown AW has a specific gravity (SG) of 0.56 kg/m^3, while the re-plantation grown AW has a lower SG of 0.40–0.45 kg/m^3 (National Research Council 1983). Specifically, In Borneo, the SG value of AW obtained could be around 0.43–0.47 kg/m^3 for a 6-year old AW (Sining 1989).

There are two types of chemicals used for the experiment; acid and alkaline. Pure acetic acid (CH_3COOH) solution was obtained from Fisher Scientific (UK), Ltd., known by the product name “Acetic Acid, ACROS Organics™”. Powdered sodium bicarbonate ($NaHCO_3$) was obtained from Fisher Scientific (UK), Ltd., known by the product name “Sodium Bicarbonate, ACROS Organics™”. Polyhydroxyalkanoates (PHA) was obtained from Jinan Pu Dong Import and Export Trading (China) Co. Ltd. with chemical abstracts service (CAS) number “718630-59-2”. PHA has a density of 1.2–1.4 kg/m^3 and contained 90% bio-based carbon according to ASTM D6866-16 (2016) standard. Pure grade powdered PLA with 100% bio-based carbon according to the ASTM D6400-12 (2012) standards was obtained from Shenzhen Esun Industrial (China) Co. Ltd., product code “Al-1001”. Pure grade powdered PLA has a density of 1.25 kg/m^3 and melt flow index (MFI) of 10–12 g/10 min. A light brown color Nano-clay hydrophilic bentonite (NCHB), with a particle size less than 25 microns was obtained from Sigma-Aldrich (US) Corporation, product code “682659”.

2.2 Fiber Preparation

Unmodified AW fiber were washed and rinsed with distilled water and was dried in an open ventilated oven at 80 °C for 24 h. The model of the ventilated oven is “ECOCELL EC55” by MMM Medcenter Einrichtungen (Germany) GmbH. The chemical treatment process involved the use of two different types of chemicals. Firstly, the AW fibers were soaked for 24 h inside a 1.5 L 5 wt% acetic acid solution (CH_3COOH) mixed with distilled water. After 24 h, 90 gm of sodium bicarbonate

($NaHCO_3$) was mixed into the acetic acid-soaked AW mixture for another 24 h. The reactions of all possible chemical are as below:

$$Na_2CO_3 + 2CH_3COOH \rightarrow 2CH_3COONa + H_2O + CO_2 \tag{1}$$

$$Na_2HCO_3 + CH_3COOH \rightarrow NaCH_3COO + H_2O + CO_2 \tag{2}$$

$$Wood - OH + CH_3COONa \rightarrow Wood - O - NaH + CH_3COO \tag{3}$$

The chemically treated AW fibers were filtered and rinsed using distilled water to neutralized remaining chemicals attached onto the fibers. The neutralized AW fibers was dried in an open ventilated oven at 80 °C for 24 h, and the dried fibers were stored at room temperature conditioning, according to ASTM E41-92 (2010).

2.3 *Fabrication of the Bio-composites*

Pure PLA, PHA, PLA/PHA (1:1 ratio of PLA/PHA), and NCHB-PLA/PHA (1 wt% of NCHB; 1:1 ratio of PLA/PHA) with different weight percentage (i.e. 5, 10, 15 and 20 wt%) are fabricated. The mixtures of the materials were blended for 5 min using a drying mixer machine. The molds used for the bio-composites fabrication, were designed according to ASTM D638-14 (2014) for tensile strength, ASTM D790-17 (2017) for flexural strength, ASTM D4812-11 (2011) for impact strength, and ASTM D150-11 (2011) for dielectric properties. Prior to the fabrication of the bio-composites, the molds were first cleaned and releasing waxed were applied before they were filled with the mixture of materials. Mold releasing wax prevents the fabricated samples from sticking to the mold and ease the removal process. Aluminum foils were placed in between the mold plates, which ease the removal process and reduced the formation of bubbles in the samples. The mixture of materials was placed in the mold as according to the different weightage, polymer and reinforcement composition. The mold was compressed using a hot press hydraulic machine model 'LS-22071' equipped by Lotus Scientific (Malaysia) Sdn. Bhd. under pressure of 1000 psi (6.89 MPa). The mold containing the composite mixture was compressed and heated in the hot press machine at 150°C for 6 min. The samples were quenched in water at room temperature, according to ASTM E41-92 (2010). The quenched samples were dried using a hair dryer and kept concealed at room temperature conditioning, according to ASTM E41-92 (2010).

2.4 Methods

2.4.1 Tensile Test

Tensile test conducted are according to ASTM D638-14 (2014) standard using Universal Testing Machine instrument, T-Machine Technology (Taiwan) Co. Ltd. model ‘UTM107’. The crosshead speed used is 5 mm/min, and tensile strengths were calculated using the formula below by dividing the maximum load by the cross-sectional area of the sample:

$$\sigma_T = \frac{F}{A_o} \tag{4}$$

where, σ_T is the tensile strength at break, F is the load, and A_o is the cross-sectional area. Tensile test was conducted for each weight percentage for five samples. The results of five samples were average for each weight percentage.

2.4.2 Flexural Test

Flexural test conducted are according to ASTM D790-17 (2017) standard using Universal Testing Machine instrument, T-Machine Technology (Taiwan) Co. Ltd. model ‘UTM107’. The crosshead speed used is 5 mm/min. Flexural strengths were calculated for each sample under a load, on a three-point bending setup, based on the equation below:

$$\sigma_F = \frac{3PL}{2bd^2} \tag{5}$$

where, σ_F is the flexural strength at break, P is the load, b is the width, and d is the depth (thickness). Flexural test was conducted for each weight percentage for five samples. The results of five samples were average for each weight percentage.

2.4.3 Impact Test

Impact tests conducted are according to ASTM D4812-11 (2011) standard using Charpy Impact Tester instrument. The machine was supplied by Lotus Scientific (Malaysia) Sdn. Bhd. model ‘LS-22 006-50J’. The angle of release was set at 120° from the sample. The results were recorded after the impact and the energy absorbed by each sample was calculated and the impact strength for 50 J energy absorbed was calculated based on the formula below:

$$\sigma_I = \frac{E}{A} \tag{6}$$

where, σ_I is the impact strength at break, E is the energy absorbed by the broken sample, and A is the cross-sectional area. Impact test was conducted for each weight percentage for five samples. The results of five samples were average for each weight percentage.

2.4.4 Scanning Electron Microscopy

Composite samples were sliced and mounted on aluminum stubs and was finely coated with metal particles for 1–2 min, depending on the size of the samples. The coating process utilize a spur coating machine supplied by JEOL (Japan) Ltd. model 'JFC-1600'.

Scanning electron microscopy (SEM) was conducted according to ASTM E2015-04 (2014) standard using Hitachi Analytical Table Top SEM (bench top) supplied by Hitachi High-Technologies Europe (Germany) GmbH model 'TM-3030'. SEM process utilizes a field emission gun with an accelerating voltage of 5 and 15 kV in order to collect images of the surface of the composite samples. Several clear microscopic images of the surface structure were obtained and discussed.

2.4.5 Light/Optical Microscopy (L/OM)

Light/optical microscopy (L/OM) was conducted according to ASTM E2015-04 (2014) standard using an industrial microscope supplied by Nikon (Japan) Corporation model 'Eclipse 3 × 2 LV140'. Several clear microscopic images of the surface structure were obtained and discussed.

2.4.6 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transforms infrared spectroscopy (FTIR) was conducted based on qualitative and quantitative analysis according to ASTM E168-16 (2016) and ASTM E1252-98 (2013) standards using FTIR Spectroscopy by Shimadzu (Japan) Corporation model 'IRAffinity-1'. FTIR used infrared spectrum of emission and absorption of the samples to create unique molecular fingerprint spectrum. Spectrum scanning was conducted within wavenumber range of 4000 cm^{-1} and 400 cm^{-1} for each sample. Spectrum scanning was conducted numerous times for each sample and the most representative results were selected and discussed.

2.4.7 Thermo-Gravimetric Analysis (TGA)

Thermo-gravimetric analysis (TGA) was conducted according to ASTM E1868-10 (2015) and ASTM E1131-08 (2014) standards using a thermal-gravimetric spectrometer supplied by Shimadzu (Japan) Corp. model ‘TA-60WS’ workstation analyzer. The analysis was conducted by heating.

2.4.8 Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry (DSC) analysis was conducted according to ASTM D3418-15 (2015) and ASTM E1269-11 (2011) standards using DSC spectrometer supplied by Perkin Elmer (USA) Corporation model ‘DSC8500 PerkinElmer Smart scan TM Instrument’. The samples were examined under flowing nitrogen (20 mL/min) between 0 and 200 °C. The calculated degree of crystallinity uses the formula as below:

$$Xc = \frac{\Delta H_m}{w \Delta H_m^0} \times 100 \tag{7}$$

Xc is the degree of crystallization, ΔH_m is enthalpy of fusion, w is the weight fraction of polymeric matrix in the composites, and $\Delta H_m^0 = 93.6$ J/g (for the 100% PLA) and/or 109 J/g (for 100% PHA) (Dagnon et al. 2009; Ero-Phillips et al. 2012; Li and Mc Carthy 1999; Wagner et al. 2014).

3 Results and Discussion

3.1 Effects of Nano-enhancement on Polymer Blend (PLA/PHA) Bio-composites Mechanical Properties

Samples tested are bio-composite Nano-clay enhanced polymer blends (PLA/PHA) and unmodified and modified AW fiber as reinforcement, Figs. 1, 2, 3, 4, 5, 6, 7 shows the results of the tensile, flexural and impact tests conducted. Figure 1 shows the tensile strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites at different fiber weight percentage. According to Fig. 1, the optimum tensile strength was achieved at 10 wt% weight percentage for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. Comparing the pure NCHB-PLA/PHA and 10 wt% U-AW-NCHB-PLA/PHA, the percentage of increment for 10 wt% U-AW-NCHB-PLA/PHA is about 8%. Between the 10 wt% samples of M-AW-NCHB-PLA/PHA and U-AW-NCHB-PLA/PHA, the percentage of increment is about 12% for 10 wt% M-AW-NCHB-PLA/PHA. Figure 2 shows the stress-strain curves of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites at 10 wt% weight percentage. Figure 3 shows the comparison of tensile strength results for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different fiber weight percentage.

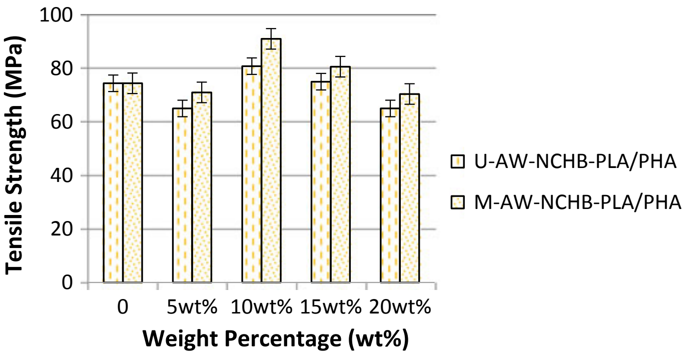

Fig. 1 Tensile strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different fiber weight percentage

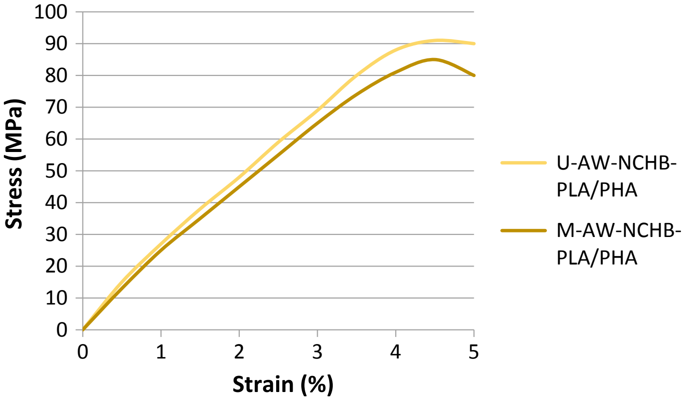

Fig. 2 Stress-strain curves of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at 10 wt% weight percentage

Figure 4 shows the flexural strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites at different weight percentage. According to Fig. 4 the optimal flexural strength for AW-NCHB-PLA/PHA bio-composites was achieved at 10 wt% weight percentage for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. Comparing to pure NCHB-PLA/PHA, the percentage of increment for 10 wt% U-AW-NCHB-PLA/PHA is about 1%. Between the 10 wt% samples of M-AW-NCHB-PLA/PHA and U-AW-NCHB-PLA/PHA, the percentage of increment is about 5% for 10 wt% M-AW-NCHB-PLA/PHA. Figure 5 shows the comparison flexural strength for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different weight percentage.

Figure 6 shows the impact strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites at different weight percentage. According to Fig. 6, the optimal impact strength for AW-NCHB-PLA/PHA bio-composites was

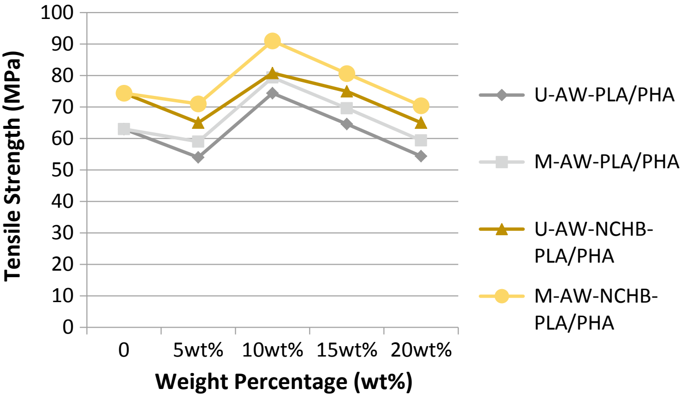

Fig. 3 Comparison of tensile strength for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different weight percentage

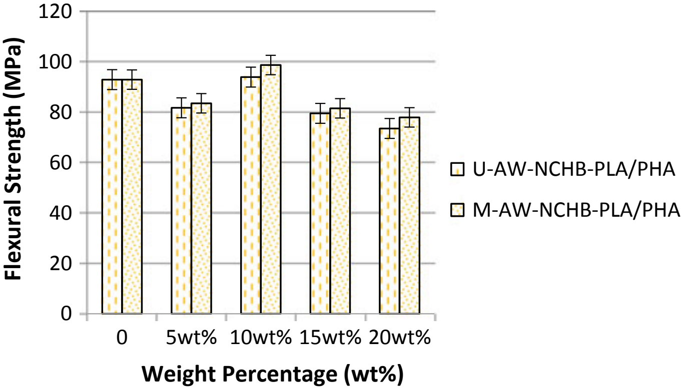

Fig. 4 Flexural strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different weight percentage

achieved at 10 wt% fiber loadings for both U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. Comparing to the pure NCHB-PLA/PHA, percentage of increment for 10 wt% U-AW-NCHB-PLA/PHA is about 21%. Between the 10 wt% samples of M-AW-NCHB-PLA/PHA and U-AW-NCHB-PLA/PHA, the percentage of increment is about 9% for 10 wt% M-AW-NCHB-PLA/PHA. Figure 7 shows the comparison impact strength for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different fiber loadings.

By incorporating NCHB in the polymer blend, mechanical properties of the bio-composite (i.e. tensile, flexural and impact strength) have improved. Nano-clay enhancement increased the viscosity of the polymer blend, hence increases the threshold mechanical strength of the bio-composites. A strong adhesion was created

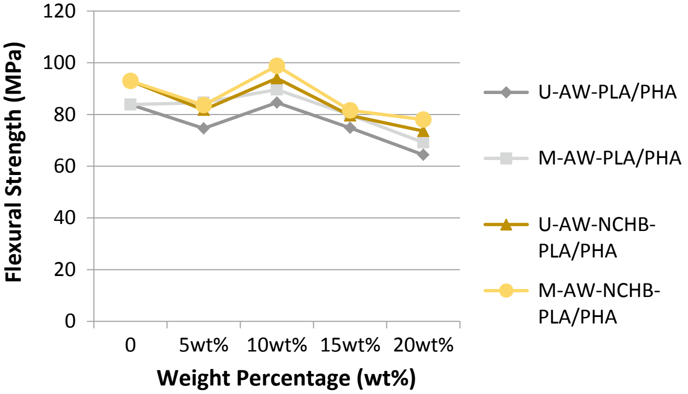

Fig. 5 Comparison of flexural strength for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different weight percentage

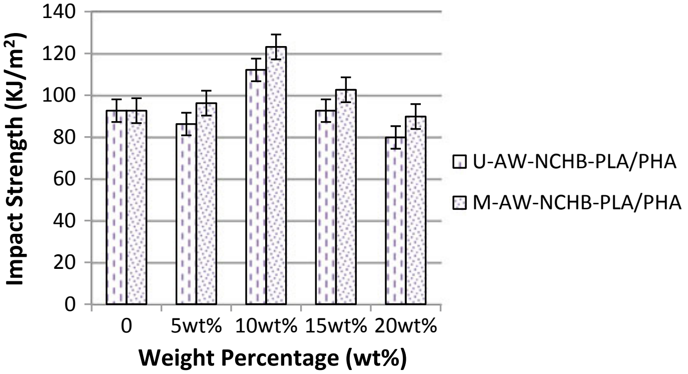

Fig. 6 Impact strength of U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different fiber loadings

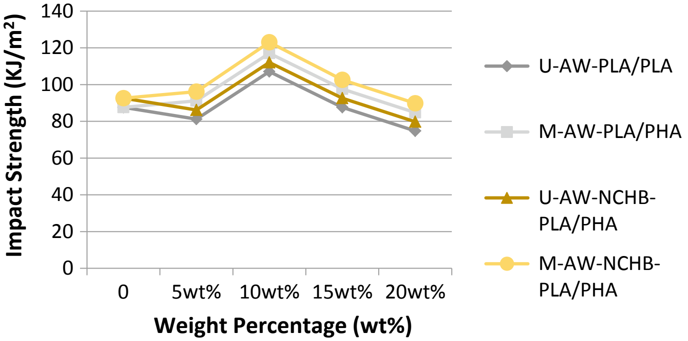

Fig. 7 Comparison of impact strength for U-AW-PLA/PHA, M-AW-PLA/PHA, U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA at different weight percentage

between fiber and polymer, due to the incorporation of Nano-clay, which reduces the agglomeration and formation of void during fabrication (Dewan et al. 2013; Haq et al. 2009). Other studies reported that, due to the size of NCHB, it could be easily incorporated, dispersed and reduces voids or air bubbles and fiber agglomeration, hence increases the adhesion between fiber and polymer (Bensalah et al. 2017; Dewan et al. 2013; Hamidi et al. 2008; Haq et al. 2009). Abdellaoui et al. (2017) also reported that due to incorporation of clay disperse in their laminated jute epoxy bio-composites, increases mechanical strength of the bio-composites compared to its non-enhancement bio-composites. Bozkurt et al. (2007) reported similar results with their samples of non-crimp glass fiber reinforced layered clay/epoxy nanocomposites. The addition of an enhancement in the composite blend increases mechanical strength when compared with composites with no enhancement.

3.2 SEM and L/OM Images of Unmodified and Modified Fiber with Nano-enhancement for Polymer Blend (PLA/PHA) Bio-composites Morphological Properties

SEM images of (a) U-AW-NCHB-PLA/PHA, and (b), (c) M-AW-NCHB-PLA/PHA, and L/OM images of (d) M-AW-NCHB-PLA/PHA bio-composites is displayed in Fig. 8. Section (a) of Fig. 8 shows a void between fiber and polymer. It was observed that the U-AW and NCHB-PLA/PHA has a smooth surface structure and could be considered as semi-brittle/semi-ductile. These properties could be affected by impurities, oily and waxy structure of U-AW fiber, hence causes poor adhesion and absorbance between U-AW fiber and NCHB-PLA/PHA. Quenching the NCHB-PLA/PHA bio-composites during fabrication causes implosion which closely packs the structure it with each other, hence influencing the polymer to be stronger with semi-ductile/semi-brittle interlocked structure. Some studies support this occurrence such as Avila-Orta et al. (2013) and Pickering et al. (2016), where controlled temperature during heating and cooling caused brittle or ductile structures to form depending on the behavior or chemical composition reaction. Due to the size of NCHB, it enables better dispersion, hence smooth surfaces were form on the bio-composites with less formation of voids (Bensalah et al. 2017; Dewan et al. 2013; Hamidi et al. 2008; Haq et al. 2009). Section (b) of Fig. 8 shows the smooth semi-brittle/semi-ductile interlocked phase of NCHB-PLA/PHA bio-composites.

Section (c) and (d) of Fig. 8 display strong adhesion between M-AW fiber and NCHB-PLA/PHA polymer due to removal of impurities such as; wax, oil and some part of the weak lignin, hemicellulose and cellulose of the AW fiber. The removal of impurities produces a rougher, porous and stronger fibril branch structure, hence influencing better interlocking between fiber and polymer which contributes towards better adhesion and improved strength of the bio-composites (Balakrishna et al. 2013). Chemical modification on the fiber created desirable wettability and absorption, which also contributed towards improved adhesion. The rougher surface, pores

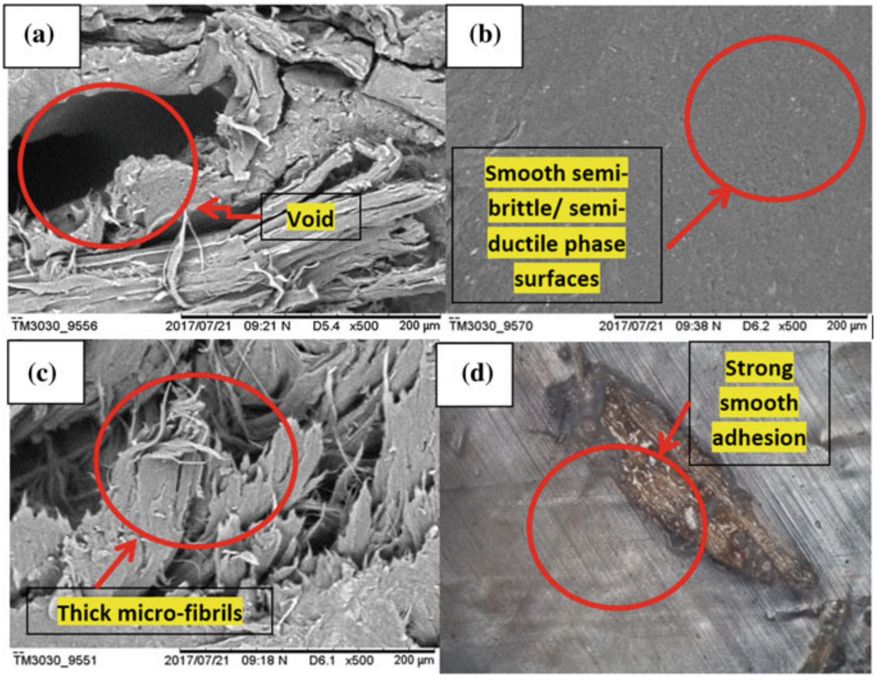

Fig. 8 SEM images of **a** U-AW-NCHB-PLA/PHA, and **b**, **c** M-AW-NCHB-PLA/PHA, and L/OM images of **d** M-AW-NCHB-PLA/PHA bio-composites

and micro-fibril branches structures also contributes higher friction which grips and secure the movement of fiber and polymer when fracture, break, load or stress were applied. According to Boynard and D'almeida (2000), securing the movement of the fiber reinforcement supports and slow down the failure of the bio-composites.

3.3 FTIR of Nano-enhancement on Polymer Blend (PLA/PHA) Bio-composites Infrared Spectral Properties

Figure 9a and b shows the FTIR spectrum of PLA/PHA and NCHB-PLA/PHA. Table 1 shows the FTIR spectrum extracted from Fig. 9. The small peak at 2935.66 and 2412.95 cm^{-1} for PLA/PHA, and 2935.66 cm^{-1} for NCHB-PLA/PHA was attributed to the C–H stretching and O–H stretching bond structure that contained functional group of alkanes and carboxylic acids. The small peak in the region of the C–H stretching bond structure could also include a functional group of methyl (CH_3), methylene (CH_2), and aliphatic saturated (CH) (Khalil et al. 2013). The reduction in one of the peaks for NCHB-PLA/PHA could be cause by the incorporation of NCHB, hence influencing a better interlocking system or adhesion between amorphous PLA and crystalline PHA (Armentano et al. 2015).

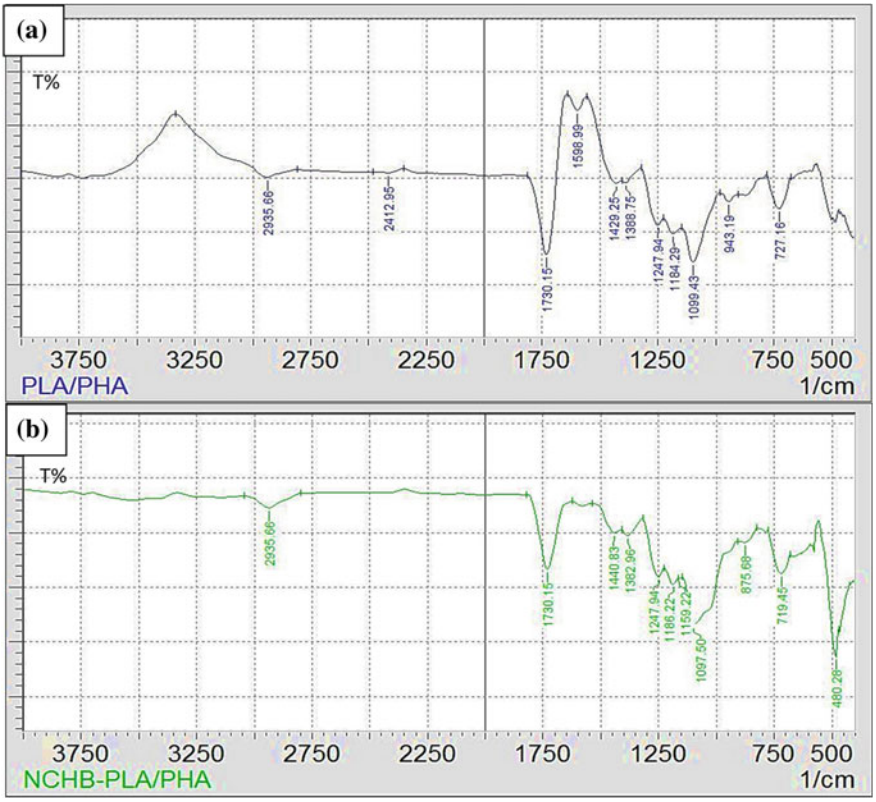

Fig. 9 FTIR spectrum **a** PLA/PHA and **b** NCHB-PLA/PHA

Table 1 The wavenumber of PLA/PHA and NCHB-PLA/PHA

Functional group	Wavenumber (cm^{-1})	
	PLA/PHA	NCHB-PLA/PHA
C–H stretching, O–H stretching—alkanes (CH; CH$_2$; CH$_3$), carboxylic acids	2935.66, 2412.95	2935.66
C=C stretching—alkenes	1730.15, 1598.99	1730.15
C–H bending—alkanes	1429.25, 1388.75, 1247.94	1440.83, 1328.96, 1247.94
C–O stretching,—alcohol, carboxylic acids, esters, ethers	1184.29, 1099.43	1186.22, 1159.22, 1097.50
C–H “oop”—aromatic	943.19, 727.16	875.68, 719.45
=CH bending—alkenes	–	480.28

The small peak at 1730.15 and 1598.99 cm^{-1} for PLA/PHA, and 1730.15 cm^{-1} for NCHB-PLA/PHA is characterized as the C=C stretching bond structure from the functional group of alkenes. This could be attributed to the mixture of semi-amorphous and semi-crystalline structure of PLA and PHA (Arrieta et al. 2015; Hu et al. 2008; Zhang and Thomas 2011) and peak at 1429.25, 1388.75 and 1247.94 cm^{-1} for PLA/PHA, and 1440.83, 1328.96 and 1247.94 cm^{-1}, for NCHB-PLA/PHA are characterized as the C-H bending bond from the functional group of alkanes (Armentano et al. 2015; Arrieta et al. 2015). The peak at 1184.29 and 1099.43 cm^{-1} for PLA/PHA, and 1186.22, 1159.22 and 1097.50 cm^{-1} for NCHB-PLA/PHA could be characterized as the C–O stretching bond structure from the functional group of alcohol, carboxylic acids, esters and ethers (Armentano et al. 2015). The peak at 480.28 cm^{-1} for NCHB-PLA/PHA could be characterized as the =CH bending bond structure from the functional group of alkenes. The peak at 943.19 and 727.16 cm^{-1} for PLA/PHA, and 875.68 and 719.45 cm^{-1} for NCHB-PLA/PHA could be characterized as the C–H "oop" bond structure from the functional group of aromatics. Few additional peaks could be cause by the NCHB enhancement, hence influencing additional free hydroxyl structure, improving adhesion of PLA and PHA (Armentano et al. 2015; Hu et al. 2008).

3.4 Thermal Crystallinity Properties of Unmodified and Modified Fiber and Nano-enhancement with Polymer Blend Bio-composites

Figure 10 shows the DSC thermogram for U-AW-NCHB-PLA/PHA and M-AW-NCHB-PLA/PHA bio-composites. Table 2 is the numerical version of the graph showed in Fig. 10. The incorporation of NCHB enhancement further reduced the

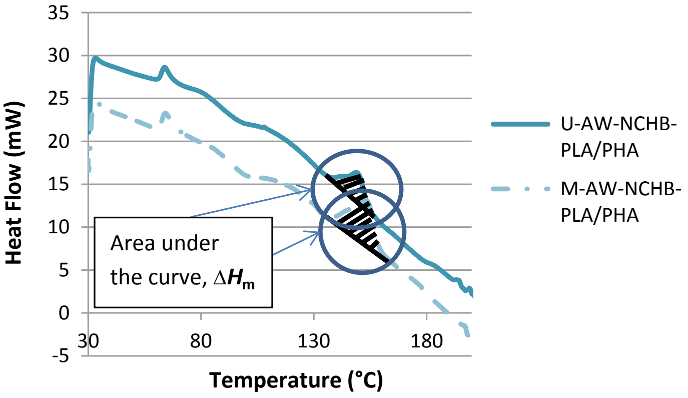

Fig. 10 DSC graph of **a** U-AW-NCHB-PLA/PHA and **b** M-AW-NCHB-PLA/PHA

Table 2 The wavenumber of PLA/PHA and NCHB-PLA/PHA

Specimen	Initial melting point (°C)	Final melting point (°C)	Peak melting temperature (°C)	Heat flow—enthalpy of fusion, ΔH_m (Jg^{-1})	Degree of crystallinity (X_c) (100% PLA)	Degree of crystallinity (X_c) (100% PHA)
U-AW-NCHB-PLA/PHA	136.0	155.5	149.2	16.3	17.41	14.9
M-AW-NCHB-PLA/PHA	136.7	162.5	154.4	12.7	13.56	11.6

Note 100% of PLA = 93.6 J/g, 100% of PHA = 109.0 J/g

crystallinity values of the bio-composites, hence influencing better adhesion or interlocking between the AW fiber and polymer blends. The crystallinity of the bio-composites was also reduced, due to the hydrogen bond formation between the modified NCHB, AW fiber and polymer blend causing the surface energy of the fiber to be closer (Kumar et al. 2010).

4 Conclusion

In conclusion, incorporating AW fiber caused changes in the morphological and functional group properties of the bio-composites. The chemical modification on the fiber causes M-AW fiber to be clean, rough, porous and more micro-fibrils branches structure to form. This due to removal of lignin, hemicellulose, wax, oil and other impurities present on the fiber. Hence, allowing more surface wettability and better interlocking, producing stronger bio-composites. Polymer blend of both PLA and PHA produces a rough interlocking structure, hence producing stronger and more flexible properties than purely PLA or PHA polymer. Nano-Clay enhancement (NCHB) promote a smoother interlocking structure between PLA and PHA polymers. This could be caused by both the Nano-size and dispersibility of NCHB, hence reducing voids which improves the bio-composites properties. It was found that PLA and PHA have semi-amorphous/semi-crystalline structure, respectively. There was an indication of lower crystallinity which promotes better adhesion between fiber and polymer. Polymer blends both improved the properties and adhesion by reducing the degree of crystallinity within the polymer structure, in addition NCHB further improves the adhesion. Modified AW fiber is more stable than the unmodified AW fiber as shown in the results for thermal stability testing. The stability of the modified AW fiber could be due to the additional hydroxyl group, which produced intermolecular bonding between fiber and polymer. It was also discovered that PLA have higher thermal stability compare to PHA due to intramolecular and intermolecular trans-esterification, hence the combination of the two polymers as a polymer blend, thermal stability increased even further. However, the addition of NCHB has minimal effect on the thermal stability due to the small amount used and was discovered to affect the dispersion and adhesion of the bio-composites. It was found that AW fiber causes an increase in the dielectric constant, dissipation factor and loss factor, this could be caused by the increased in the orientation and interface of the fiber, and electronic and atomic polarization in the bio-composites. The chemically modified AW fiber causes a decrease in the orientation polarization, hence increases the hydrophobicity of the modified AW fiber. The incorporation of NCHB enhancement assist in stabilizing the dielectric constant, dissipation factor and loss factor, hence leading towards obtaining smoother/consistent pattern data, in comparison with other results. The smooth surface structure could also be an indicator due to addition of NCHB in the bio-composites.

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Possibility Usage of Acacia Wood Bio-composites in Application and Appliances

A. Muhammad, Md Rezaur Rahman, S. Hamdan and J. Ervina

Abstract The factors affecting the mechanical and physical properties of these materials were identified and deliberated. It was found that Acacia wood has the prospective to be efficiently produced and used in Borneo. Acacia wood has been found to be having variety of applications, among which include fire breakers, furniture production, fiber cement, doors, automobile parts, timber resource and as reinforced materials in other composites. Of significance recently is creating awareness and legislations towards producing completely biodegradable products, sustainable, environmentally friendly, durable and high strength to weight ratio. These properties are found in composites made of acacia wood, hence their potentials in wide varieties of applications, which this chapter seek to highlight.

Keywords Nano-enhancement · Mechanical strength · Infrared spectrum · Thermal properties

1 Introduction

The application of acacia wood composite has been on for many decades all over the world. acacia wood has find applications in building and construction, automobile, upholstery and furniture industries (Cogswell et al. 2016). Attention of researchers and scientists have been extensively focused on investigation of natural fiber reinforced composites. Fibers such as jute, bamboo, flax, sisal, and pineapple, wood flour, kenaf, are presently being exploited as reinforcing resources in plastics, which have light weight, low-cost and with good mechanical and thermal capabilities for wide variety of applications (Baillie 2004). Recyclability and biodegradability have made utilization of wood fiber composites a major advantage (Mitra 2014).

In contrast to synthetic fibers, it could be predisposed of upon the conclusion of their life cycle by composting or by the retrieval of their calorific worth which is not possible with synthetic fiber such as glass fiber (Begum and Islam 2013). Figure 1

A. Muhammad · Md R. Rahman (✉) · S. Hamdan · J. Ervina
Faculty of Engineering, Universiti Malaysia Sarawak, Jalan Datuk Mohammad Musa, 94300 Kota Samarahan, Sarawak, Malaysia
e-mail: rmrezaur@unimas.my

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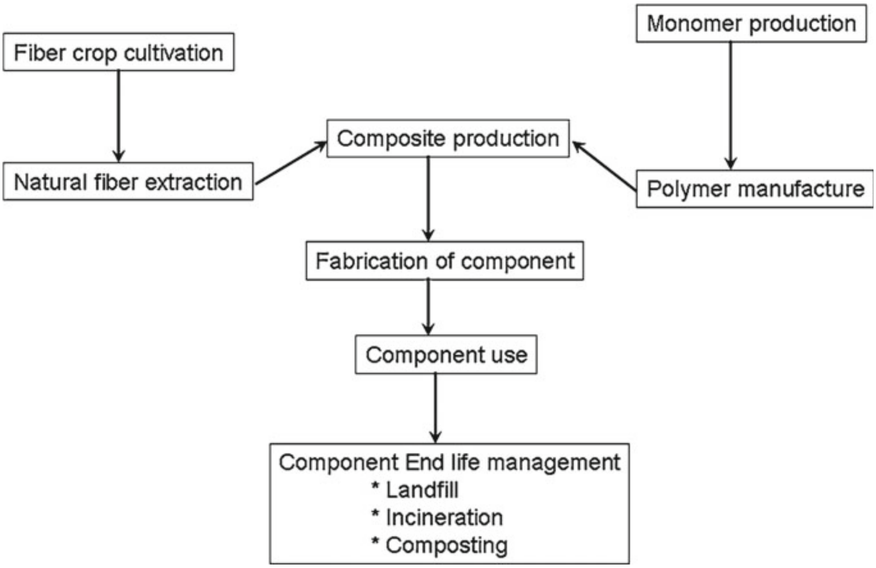

Fig. 1 Life cycle of a composite material

indicates the life cycle of a composite material. Though, a major disadvantage of wood composites including acacia/wood flour filled composites is that they are polar in nature which makes have hydrophilic characteristics, that is, they absorb water easily (Sathishkumar et al. 2013).

To address such difficulties, surface modification process of the fiber surfaces through established methods such as cold plasma treatment and corona treatment and chemical modification methods like as alkali treatment, benzoylation dewaxing and cyanoethylation are employed (Muhammad et al. 2019). Several years, wood has been employed by plastic industry as cheap filler to enhance strength and stiffness of plastic materials because of its light weight, affordability, availability and bio-based nature (Ramage et al. 2017). Waste management challenges, ecological issues and high cost are the most established contentions for the replacement of synthetic fibers such as glass fibers with wood fibers (Ramesh et al. 2013). Wood fibers exhibits mechanical properties in composites that can meet up or sometimes even be better, decreases weight of product by about 15% and have enormously receptive surface structure on the other hand causes health issues such as skin and respiratory issues which can be mitigated if not eliminated by the utilization of wood composites, chips and flakes are example of wood particles been employed as reinforcement materials in composites. Progress is being made currently in the use of wasted wood materials are and then changed over into useful items, and this pattern will in a similar vein be continually exploited. It has been reported that about 5–11% of log of wood becomes waste in form of sawdust of chips during the period of processing (Sobczak et al. 2012).

Acacia wood polymer composites including wood flour polymeric composite products have produced another class of materials that can be used as substitutes for solid wood and composites products made from synthetic based sources for diverse of applications, due to its good mechanical properties and performance, high robustness properties, low upkeep, thermal stability and wear/abrasion resistances (Stark 2003). Acacia wood has great potential as reinforcing filler owing to its easily cost benefit, availability, strength, low weight, and reasonable isotropic consistency in properties when compared to other lignocellulose materials used as fillers (Ansell 2015).

Acacia wood fibers as reinforcement in composites gives viable advantages compared to synthetic fibers. The important features which qualify fibers as reinforcement in composites are low density, strength and stiffness (Sathishkumar et al. 2014). These properties are dependent on the structure and the specialized modifications techniques because the wood fibers variables are not easily controllable as far as such qualities are applied (Huda et al. 2006).

To delve into the applications of acacia wood composites, it is important to understand that the commonly large values for strength and stiffness of lignocellulose fibers depend largely on the natural variation of plant type and its source, circumstances of its growth, and harvesting processes, and moderately its early stage when make available for the industrial usage as reinforcement in composites (Iwamoto and Endo 2015).

Over the last few decades, several applications of thermosetting and thermoplastic polymer composites have been on the increase with the combination of variety of reinforcing filler fibers (Sudhakara et al. 2013). Wood flour is a by-product of cutting, pounding, penetrating, sanding, or generally pummelling wood with a saw or other instrument (Hosseinaei et al. 2012). However, the utilization of wood-based fillers is not as prevalent as the utilization of organic fibers, yet wood based fillers have advantage and of interest over normal fillers and reinforcing agents which includes low density, flexibility during processing with less effect to the equipment, acceptable specific strength properties and low cost per volume basis (Ayrilmis et al. 2015).

From Fig. 2, it can be observed that, there are extensive usage of wood composite in automotive consumers closely followed by construction and then in marine

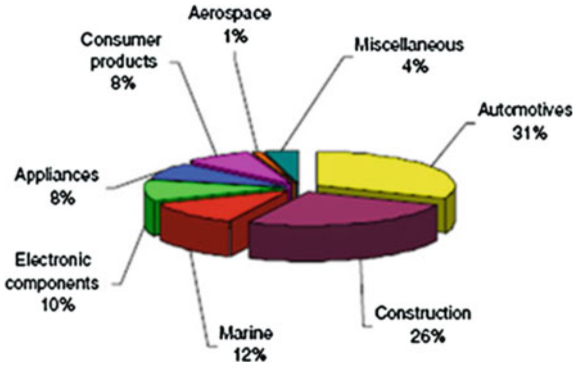

Fig. 2 Distributive applications of wood

applications. This tremendous share of wood polymer composites in automotive is of greater concern for environmental protection and the recent stiff guidelines which stipulates that 85% of the weight of vehicle has to be recyclable (Tian and Chen 2014).

2 Acacia Wood Bio-composites in Application and Appliances

2.1 Panel Production for Construction Industry

Americas has been known for utilization of residue fiber (bagasse) from the sugar cane processing for production of biocomposites (Boontima et al. 2015). In fact, the first bagasse biocomposite was manufactured in 1920 (Balaji et al. 2015; Anggono et al. 2017). There is abundant bagasse all over North America which is composed of pith and fiber. They are used for particleboards, fiberboards, and composition panel production in North America. Venezuela were the first manufacturer of bagasse fireboards in 1958 (Sant'Anna et al. 2014).

Apart from bagasse, other agriculture fibers have also been used as composites in different applications. For example, in Peru, bamboo and acacia wood fibers have been used in the fabrication panels in the construction industry (Reddy and Yang 2005) (Sanadi et al. 1995). This kind of construction usually involves the use of low technology in the prefabrication of panels shown in Fig. 3. Similarly, the north

Fig. 3 Wood composite panel

western region of united states, wheat and ryegrass straw fibers have utilized to produce panels (Youngquist et al. 1993) while in Minnesota, acacia wood stalks and hulls have been used for panel applications (Griffin et al. 2011).

2.2 Acacia Bio-Composites in Building Construction

Bio-composites from acacia wood have been have found be gradually used in the housing and construction industry. For example, it has been used in production of building compartments such as fencing, decking, siding, door, window, bridge, fiber cement, etc. These are discussed below.

2.3 Fiber Cement Composites

Fiber cement is a versatile acacia wood product utilized in the building industry (Ardanuy et al. 2015; Nakagawa and Akihama 2004). It is a fiber reinforced composite developed. One of the main materials in building products is fiber cement that has found its most versatile and increasing uses. Fiber cement composite products have largely been used the exterior and interior of buildings such as roofing, external cladding, internal lining, floors, walls, building boards, bricks, bracing, fencing, and decorative elements (Zollo 2002; Silva et al. 2010). Fiber cement is also used in construction works such as dam, bridge deck, road building, side walk, flagstone paving, and etc. (Pehanich et al. 2004).

It is important to note that one of the main ingredients of fiber cement composite are cellulose sourced from acacia wood (Semple and Evans 2007). There is also addition of chemical additives to reinforce the composites for non-structural productions specifically for making thin sheet products and fiber cement siding materials (Hadi et al. 2012). For example, as shown in Fig. 4, recently there have been production of commercial acacia wood cement reinforced composite for making panel and lap siding, simulated cedar shake and shingle products, cladding, and slate roofing, tile backer board underlayment, architectural elements, lumber substitutes such as trim, soffit, fascia, and corner boards (Evans 2002; Kim et al. 2009; Wolfe and Gjinolli 1996). Acacia Wood fiber cement composites have also been produced and has remain a vital substitute for asbestos in building and construction industry (Kim et al. 2011). Very importantly, there is complete elimination of problems of rots and insects’ attacks from acacia wood cement composite products and they also have high fire resistance with better durability (Sakagami et al. 2009; Semple and Evans 2002).

Fig. 4 Product of fiber cement composites

2.4 Exterior Construction in Building

The application of acacia wood composites has been used in the construction of exteriors in the building industry. Acacia wood composite are used to make standard lumber in a different cross-sectional dimension (Wickens et al. 1995). These products are utilized in the production of boards, deck, picnic tables, landscape timbers, and industrial flooring (Anoop et al. 2012). Lumbered products from acacia wood composites are also recommended by manufacturers for applications in biocomposites needed in the construction of gaps on the edges and ends of a thermal expansion. This construction gap also serves as composites that check moisture absorption (Midgley and Turnbull 2003).

2.5 Window and Door Construction

Solid Acacia wood composites have been utilized in the making clad components for doors, windows (Missio et al. 2018). It has been reported that application of solid wood alone needs broad cutting, gluing, finger-jointing and edge for clearance of window and door sections during their fabrication (Çetinkol et al. 2012). Similarly, the glued section needs to be milled for accurate cross sectional area assembly and these consequently increase the construction cost and a waste of wood components (Grasel et al. 2015). Because of these many disadvantages associated with solid wood, manufacturers have turned to wood composites to produce window and door components. Figure 5 shows doors made from wood composites.

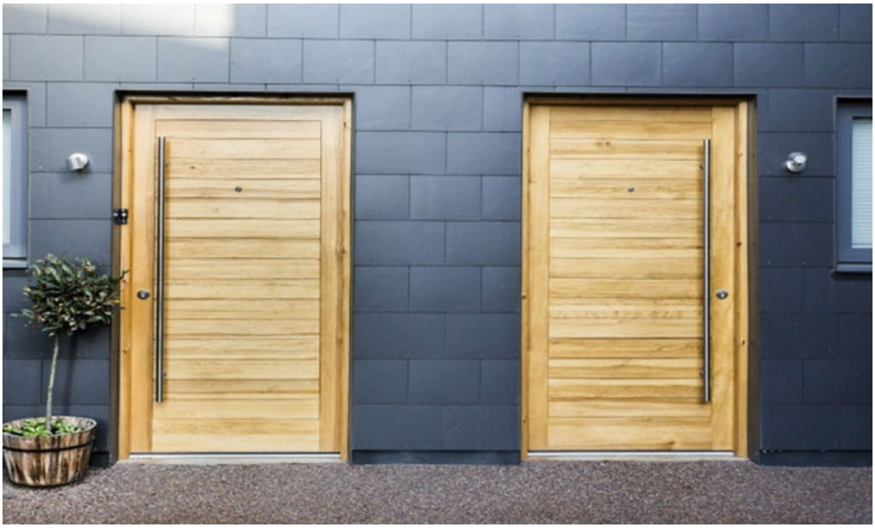

Fig. 5 Acacia wood composite doors

2.6 *The Green Kitchen Construction*

Recently, Acacia wood composites have been utilized in the building of green kitchen. In the construction of green kitchen, renewable wood-based materials composites are replacing the finite petroleum-based systems to reduce green gas emissions. Figure 5 depicts complete set of appliances in kitchen construction from wood. Some of the products of Green kitchen are.

2.6.1 Carpets

Acacia wood produced from corn has shown to be flexible components that mitigates many enzyme processes and has been used to build bio products. For example, the polyactic acid (PLA) produced from acacia wood corn composites has been used to make carpet (Arthanarieswaran et al. 2016). In addition, it has been revealed that carpets produced have unique properties of breaking down under composting conditions (bin Bakri et al. 2018).

2.6.2 Cupboard

Cupboard can be made from acacia wood/straw/rice fibres through production of polyurethane resin. The resin fiber straw is bound to the board through pressing or

Fig. 6 Kitchen cabinet constructed from acacia wood

it can be laminated or stained to the wood for more reinforcement. The cupboard produced has been found to be environmentally friendly as the by-product that needed to be burned after every growing season can be utilized (Taflick et al. 2015) (Fig. 6).

2.6.3 Countertop

Acacia wood fibers composites has been used for injection molds to cast countertops that are uncommonly light and durable because the strength of the top can cannot easily be broken unless by bacterial due polyurethane that harden the mass. In general, countertop is interesting due to its biodegradability characteristics (Shebani et al. 2009).

2.7 Bridge Construction

Acacia wood composites has been utilized in the production of Stay-in-place (SIP) bridges and distance between bridge girders. It has been revealed that SIP bridges made from acacia wood fiber composites have various benefits than that made from steel because they are breathable and porous (Dubey et al. 2009). The porosity of the composite allows water to evaporate through form and this prevent corrosion. The composite is also biodegradable with a potentiality of breaking after long time us and this also allows for inspection of the bridge deck. It is also very light when compared with steel and this enhances faster and cheaper installations (Senthamaraikannan et al. 2016).

2.8 Precast Fibrous Concrete Panel

Acacia bio-fibers composites have been used for the fibrous reinforcement during the precasting of concrete panels (Kull et al. 2011). Usually, the precasting is done to stabilize the ground slope especially in areas close to the riverbanks. It has been reported that the application of reinforced precast fiber composite also reduces the amount of steel reinforcement which further improves the shear and bending capabilities of the elements and promotes green building initiatives (Castro-Díez et al. 2011).

2.9 Acacia Wood Products

2.9.1 Fuel

One of the major applications of acacia wood is its utilization as fuel in the most rural communities (Kumar and Gupta 1994). It is important to note that this application is exceeding the required incremental growth because the annual volume of Acacia wood usage over the last twenty years is 8.5 m^3/ha which equivalent to 0.42 m^3/ha of mean annual increment (Marsoem and Irawati 2016). The production of charcoal has helped in overcoming the problems of transportation. Charcoal is used to provide energy for cooking and for production of electricity (Eriksson et al. 2003). Firewood is another vital product for acacia wood species. Its production is premise on the its ability to grow rapidly when cut requiring little amount of energy and water, easy to handle and only few thorns and can readily be transported. It is very important to note that Charcoal, is much desire in the urban area. Its manufacture is therefore very lucrative though there is low output efficiency due to ineffective practices involves in its production process (Ahmed et al. 2018). It has been reported that the inefficiency can be as low as 15% which consequently made it wasteful process (Souza-Alonso et al. 2017). Firewood from acacia species are slow in burning with high calorific content and with little or no smoke. The fumes it produced is also nontoxic and it does not spark during burning (Fagg and Stewart 1994).

Another important application of acacia wood composite is in automobile dashboard components. A lot of investment is done by large companies in utilization of properties of acacia wood in this sector. Figure 7 shows how the wood is used in the design of the interior of most cars recently.

Fig. 7 Automobile parts made from acacia wood composite

3 Non-wood Products

Acacia wood species are being used in non-wood products.

3.1 Forage

Forage production from acacia wood has been found to be non-toxic, nutritious fruity. It can also withstand browsing, pruning, lopping and coppicing. Similarly, they are quick growing especially at early stage (Carlmark 2013). They are herbaceous and readily available to livestock and game animals. They are often dried and cured plant. Similarly, acacia trees also provide valuable browse for both game and livestock as well as being a valuable source of highly nutritious pods which can be stored as a dry season supplement for livestock. Asides this, the woody part of the acacia plant also provides high quality food source for undulates at the critical period of their pregnancy (Strong and Gates 2009). The utilization of acacia wood plant as a source of forage is not only limited to herbivores, it is also good source of pollen grains for bee production. For example, while wild honey can be collected from natural cavities of trees (Ralph and Hatfield 1991).

3.2 Gum

The utilization of acacia wood polysaccharide species has been seen in different domestic products such as adhesives, ink constituents, cosmetics, confectionery as food. in crafts, as a cosmetic, in confectionery and as a food (De Simón et al. 2014). Gums from acacia species are also used in production of traditional medicine which are used as soothing agents to combat diseases such as coughs, diarrhea, dysentery, hemorrhage, and external coverage of inflamed areas on the skin (Alamgir 2017). It is worth to note Acacia wood specie from Senegal and 18 other species of this brand have found used for commercial and industrial applications especially pharmacology (Behera and Dhir 2013).

3.3 Tannins

The acacia wood species that has bark or pod have been utilized as a source of tannins for the leather industry (Ping et al. 2011). Tannins are vital ingredients used in the tanning industry for production of finished leather goods. The exploitation of tannins from Acacia species has been recorded in many locations in sub-Saharan Africa countries such as Senegal, Mauritania and Mali (Ping et al. 2011). The residues from the pods of these species contain up to 50–60% tannin, which is used when used in tanning gives a soft, plump, very light-colored leather (Jordão et al. 2017).

3.4 Fiber

The root and stem of acacia wood species used for production of fibers which are vital in the domestic economy with commercial potentials. The fibers are essential constituents for production composites (Ingleby et al. 1997).

3.5 Local Medicine

The acacia wood species has found applications in the production local medicinal products for non-clinical studies and treatment of venereal diseases such as diabetes or as aphrodisiac etc. It is important to note that their usage is problematic since there is efficacy issues. For instance, Gum Acacia specie has been used as an emollient anesthetic that result in softening of tissues on the skin and irritated internal surfaces (Rajvaidhya et al. 2012).

3.6 Food

The seeds of several African acacia wood species were known to be consumed as food either chewed, cooked or raw, though very little has been known about the nutritional content (Carruthers et al. 2011). Though, it has been reported that some Australian species of Acacia wood species are found to be rich in protein containing 17–25%, while the fatty content is about 41.6% and carbohydrate is about 30–40% (Miller et al. 2011). These values have been found to compete favorably with the ones found in cereals, wheats and rice and even found to higher than those found in meats.

3.7 Handicrafts

Acacia wood species has been used productions of handicrafts. They have found application in small commercial quantity in the tourist industry (Dogan et al. 2008).

4 Other Essential Services of Acacia Wood

4.1 Environmental Protection

The acacia wood had been used for protection of the environment because those found in the drier regions have extensive, subsurface lateral roots systems that has the capacity to take advantage of light rain rainfall that helps in stabilizing the soil (Ramage et al. 2017). The nature of the aerial system coupled with extensive root system also helps in reducing wind erosion and ameliorate the possibility of microclimate change information (Cornwell et al. 2009). Similarly, the shade provided by the acacia wood reduces the air temperature and soil evaporation that discourages desertification. It is also important to note that low shade temperature also enhances the more nutritious ground cover even as the rainfall collected due to stem flow is directly available to the lateral roots (Kumar and Gupta 1992).

4.2 Soil Fertility

The contribution of Acacia wood species to soil fertility has been reported to be twofold. First is the nitrogen fixation potential secondly being the litter leaf fall. The contribution from leaf fall is usually due to recycling of the minerals extracted for the soil by the rooting system of acacia wood species. Though, it has been reported that the contribution by nitrogen fixation is very minimal due to the foliage on the ground level because of rapid degeneration (Nambiar and Harwood 2014).

4.3 Hedges, Shade and Shelter Usages

Acacia wood species have been used for live and brushwood hedges. The shade from the trees prove shade and shelter for livestock especially during the midday heat period. The practice of using brushwood has been extensively used by agriculturalist to make the livestock move away from the cultivated lands (Martínez-Gil et al. 2018).

4.4 Wildlife Resource

Acacia wood species has also been recognized to attract mammals, birds and edible insects due to present of large herbivores. This very important production of herbivores has made the management of game reserve feasible and has also guard against over grazing. There is balancing of the ecosystem since difference species of prey and predators are maintained and put under control (Slik et al. 2008).

4.5 Amenity and Ornamental Value

Acacia wood species have reported to be attractive for making ornaments. Though this usage has been hampered by the relatively slow growth of wood species and this has made this very application unpopular (Moon et al. 2011).

4.6 Agroforestry

Acacia wood species had found wild application in the agroforestry in which large number of their species are collected for preservation of ecosystem (Kuilla et al. 2010). This practice has been adopted by the traditional system whereby a large piece of land is cultivated so as to maintain the fertility of the soil and conservation wood species. In the long run, this conservation effort usually provides a veritable source of fuel, pole timber, herbal medicine also food for wild animals. It has also been reported the conservation of acacia wood species also influences the water imbalance right from the inception rainfall through evapotranspiration and water infiltration in the soil. This bring about rainfall redistribution and eventual recycling of materials as well as the soil fertility (Gao 2012).

5 Conclusion

As the quality of acacia wood becomes better and the prices more attractive, there will be further extension of the range of products among which include, coffered decking produced by injection molding, fixing materials, strips, underlayment, etc. Also, the door is still open for further building applications facades and fence systems. The automotive business offers other potentials, especially for the big and established manufacturers with top qualities and long-term assured distribution channels. The furniture construction sector has more to offer and having a very big potential. Acacia wood would also find numerous applications in the developments of consumer goods sector, not only in outdoor products, office supplies, advertising materials, flowerpots, but also in packing materials and pallets. Also, in the future, other mass-produced articles, toys and disposable products are expected.

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