

A BRIEF REVIEW ON THE UTILIZATION OF BIOPOLYMERS IN THE MANUFACTURING OF NATURAL FIBER COMPOSITES

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Abstract

The study briefly reviews the potential of biopolymers such as polylactic acid (PLA) and Polyhydroxyalkanoate (PHA) to become a green matrix in developing entirely biodegradable composites. The suitability of PLA and PHA in the production of entirely biodegradable natural fibre composites is discussed in this paper. The thermal properties investigation reveals that PLA and PHA are compatible with natural fibre for biocomposite fabrication. Furthermore, this study investigates the effect and performance of mechanical properties, predominantly tensile properties of combination between different natural fibres with biopolymers. In Addition, all essential elements affecting the mechanical characteristics of biocomposites are highlighted. Therefore, the current study's findings are expected capable to provide a clear picture of the biopolymer's position in producing biocomposites. It is also expected that the findings from this study will help further improve the performance of natural fibre reinforced biopolymer composites.

Keywords: Biopolymer, Natural fibres, Natural fibre composites, Polylactic acid, Polyhydroxyalkanoate, Sustainable development goal.

1. Introduction

The increased environmental awareness in society has made natural fibre composite very attractive for consumer and engineering applications. It is due to its biodegradable characteristics [1-4]. However, the current fabrication of natural fibre composites is still dependent on petroleum-based polymers as matrices such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), high impact polystyrene (HIPS), nylon, polycarbonate (PC), polypropylene (PP) and polyvinyl chloride (PVC) [5, 6]. This phenomenon produced only partially biodegradable material. Therefore, the innovations of biopolymers provide a solution as a green matrix in the fabrication of entirely biodegradable natural fibre composites.

According to the European Bioplastics Association, biopolymers must exhibit two essential characteristics, which are renewable and biodegradable [7, 8]. Renewable sources based on polymers are not always biodegradable. Biodegradability is directly proportional to the material's chemical structure, not the polymer's origin. According to ASTM D5488-94d, degradation is the ability of a polymer to decompose into simple molecules found in the environment, such as carbon dioxide or biomass, due to the enzymatic action of microbes. This process can be quantified using standard testing over specified periods [9]. Certain polymers derived from natural sources may lose their biodegradability due to chemical alteration, such as Nylon 9 polymers derived from oleic acid monomer polymerization or Polyamide 11 polymers derived from castor oil monomer polymerization [10]. Meanwhile, several petroleum-based polymers, such as polybutylene adipate terephthalate (PBAT), are biodegradable.

The availability of a biopolymer in the current market is still limited. Currently, biopolymers represent less than one percent of more than 367 million tons of annual polymers production. However, market developments for biopolymers continued to increase positively compared to the global polymer market pattern, which slightly declined. Additionally, according to the most recent market statistics produced by European Bioplastics in partnership with the nova-Institute, biopolymer manufacturing capacity is expected to expand from around 2.42 million tonnes in 2021 to approximately 7.59 million tonnes in 2026 [11].

Poly(lactic acid) (PLA) is the most promising biopolymer used as a matrix in producing biocomposites [12, 13]. On the other hand, Poly(hydroxyalkanoate) (PHA) is also beginning to be given attention by researchers for use as a matrix in developing entirely renewable and biodegradable natural fibre composites. From 2011 to 2021, Fig. 1 illustrates a considerable increase in research articles published on using biopolymers to fabricate natural fibre composites. Surprisingly, the number of research articles published on natural fibre-reinforced PLA composites has surged by 567 percent during the last decade. Meanwhile, the research articles published on natural fibre reinforced PHA composites has surged by 389 percent. This situation demonstrates that the use of biopolymers like PLA and PHA in the fabrication of natural fibre composites has garnered considerable attention and interest in the industrial, commercial, and educational sectors, as well as in research and development.

The suitability of PLA and PHA in the production of entirely biodegradable natural fibre composites are discussed in this paper. In addition, the impact on the mechanical properties of natural fibre composites by using PLA and PHA as a matrix was discussed. As a result, the current study's findings should provide a clear

picture of the biopolymer's position in producing entirely renewable and biodegradable natural fibre composites.

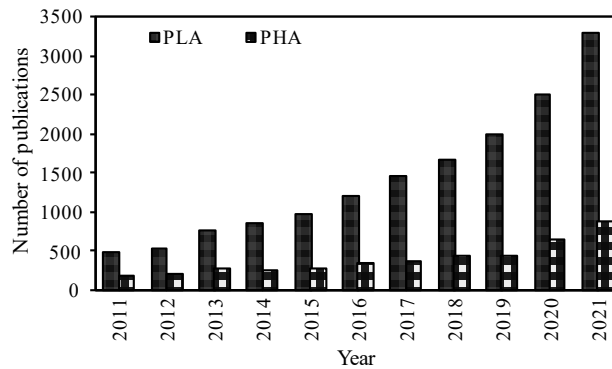


Fig. 1. Articles publication according to ScienceDirect.

2. Biopolymers

Presently, biopolymers can be classified into two main groups according to their production process, which is through the polymerization process or natural biology process. The first method in producing a biopolymer is a polymerization process, like producing PLA [8, 14]. Meanwhile, the second process in producing a biopolymer is the natural biological process through micro-organism reaction, like producing PHA.

2.1. Polylactic acid (PLA)

Polylactic acid (PLA) is a linear aliphatic thermoplastic polyester made entirely of renewable resources such as sugar, corn, potatoes, and cane [15]. The most popular industrial technique for producing high molecular weight PLA is a sophisticated controlled ring-opening polymerization (ROP) using lactide monomer derived from lactic acid, created through the fermentation of renewable agricultural resources [16].

The world's leading PLA producer is NatureWorks, which operates the world's first PLA biopolymer plant in Blair, Nebraska, United States. Its current production capacity is 150,000 tonnes per year [17]. In addition, a new NatureWorks plant will be built through a joint venture between Cargill and GC International, a wholly-owned subsidiary of Thai government-controlled petrochemical producer PTT in Thailand's Nakhon Sawan province. With a capacity of 75,000 tonnes per year, the plant will be the world's first PLA biopolymer plant to fully integrated and is expected to be operational by 2024 [18, 19].

The world's other major PLA producer is Total Corbion which operates a plant with a capacity of 75,000 tonnes per year in Rayong, Thailand. The plant began its operations in December 2018. The company is a joint venture between Total and Corbion. In addition, the firm also plans to build another plant in Grandpuits, France, with a production capacity of 100,000 tonnes per year and is expected to be operational in 2024 [20].

Interestingly, the thermal properties of PLA show good compatibility with natural fibres due to its melting temperature of 160 °C. The majority of petroleum-based polymers have a melting temperature higher than 200 °C. Meanwhile, most of the natural fibres have a degradation temperature of 200 °C [21, 22]. Therefore, a melting temperature of 160 °C makes PLA can be fabricated through a biocomposite manufacturing process before the natural fibres are degraded or decomposed.

In addition, PLA has competitive mechanical properties compared to current petroleum-based polymers, where average tensile and flexural strength is 53 MPa and 80 MPa, respectively, as presented in Fig. 2.

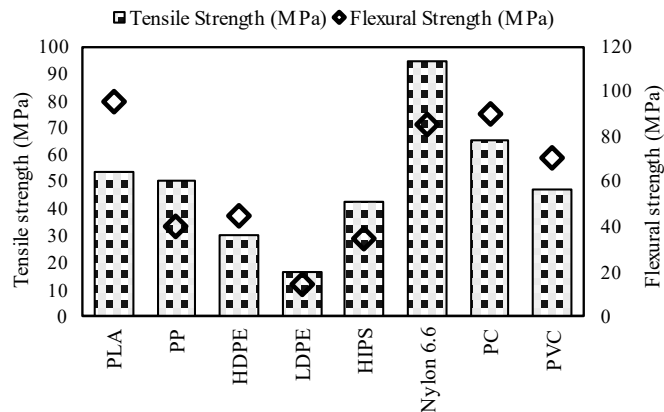


Fig. 2. The comparison of tensile strength and flexural between PLA and selected conventional polymers [23, 24].

According to the current trend in the PLA developments and its properties compatibility, PLA appears to be the potential candidate as a biopolymer in replacing petroleum-based polymers to develop sustainable, fully renewable and biodegradable natural fibre composites.

2.2. Polyhydroxyalkanoate (PHA)

The PHAs are biogenic polyesters produced in nature by numerous microorganisms, including through bacterial fermentation of sugars or lipids [25]. The PHA polymers are usually produced by bacteria grown on agricultural raw materials. PHA belongs to alkanates class obtained through bio-refining for plastic production [26]. More than 150 monomers can be combined within this family to give materials with extremely different properties. Poly (3-hydroxybutyrate) (PHB) is the most widely used and best-known PHA. PHB is a linear polyester of D(-)-3-hydroxybutyric acid that was first discovered in bacteria by Lemoigne in 1925 [27]. Also, other PHA types are Poly(3-hydroxybutyrate-co-3-hydroxyvalerate), commonly known as PHBV, polyhydroxyhexanoate (PHH) and polyhydroxyoctanoate (PHO) and polyhydroxyvalerate (PHV).

World production of PHA increased from 5.3 million tons to 17.0 million tons between years 2013 and 2020. The top world PHA producers are Danimer Scientific Bainbridge, PhaBuilder Beijing, Shenzhen Ecomann Biotechnology Co. Ltd. and Tianjin GreenBio Materials Co. Ltd. with a minimum production capacity of 10,000

tonnes per year for each company [25]. Other than these four companies, ten more companies are producing PHA in the world with a production capacity between 0.25 tonnes per year to 10,000 tonnes per year. Shenzhen Ecomann Biotechnology Co. Ltd., China is expected to increase the PHA production capacity to 75,000 tonnes per year in the next few years. Therefore, PHA also appears to be the suitable candidate of biopolymers in replacing petroleum-based polymer, especially for the fabrication of entirely renewable and biodegradable natural fibre composites.

In addition, same as PLA, the thermal properties of PHA show good compatibility with natural fibres due to its melting temperature of 160 °C ~ 175 °C [28]. Therefore, the melting temperature of 160 °C ~ 175 °C for PHA makes the PHA fabricable through the manufacturing process of biocomposites before the natural fibre degrades or decomposes as shown in Fig. 3.

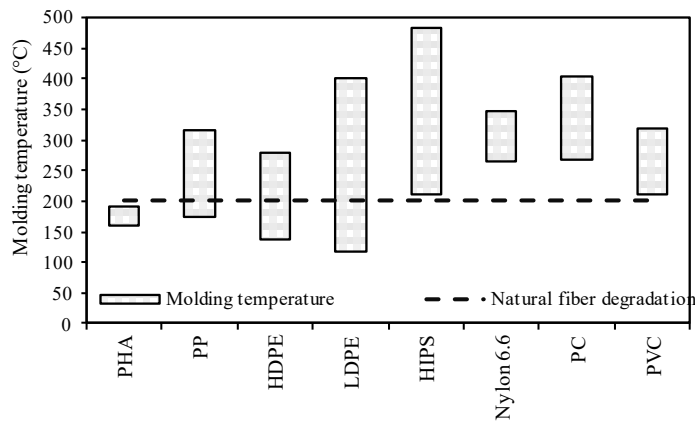


Fig. 3. Comparison of moulding temperature range between PHA and petroleum-based polymer [29-32].

In addition, PHA also has competitive mechanical properties compared to current petroleum-based polymers, where tensile strength and flexural strength is 26 MPa and 16.7 MPa respectively, as presented in Fig. 4.

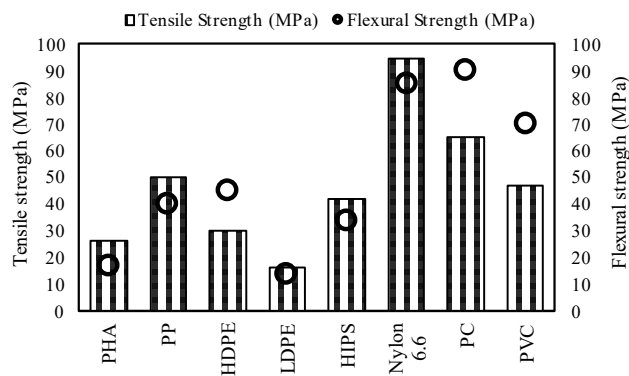


Fig. 4. The comparison of tensile strength and flexural between PHA and selected conventional polymers [23, 24, 33].

According to the current trend in the PHA developments and its properties compatibility, PHA also appears to be potential candidate as an alternative to petroleum-based polymers in developing sustainable, fully renewable and biodegradable natural fibre composites.

3. Natural Fiber Reinforced Biopolymer Composites

Natural fibres can be obtained from two main groups: plants and animals [34, 35]. The first category of natural fibre from an animal is silk, fur and hair. Meanwhile, plant or lignocellulose fibres have seven main types, depending on the plant's extraction part, such as stem, leaf, seed, fruit, root, stalk and grass [36]. However, natural fibres from plants dominate the development of natural fibre-reinforced biopolymer composites.

On the other hand, composite materials can also be classified according to the type of fibre, either continuous or short fibre-reinforced composite. However, short fibre composites have gained considerable attention because of their processing advantages, anisotropy properties, and low cost [37]. In addition, George et al. in their study presented that short fibre is better in distribution, dispersion and alignment along the direction of flow than a longer fibre length [38]. This condition results in better mechanical properties such as tensile and flexural strength. Therefore, this study investigates and analyses the potential and impact of using short natural fibres from plants for reinforcement in PLA and PHA composites.

3.1. Natural fibre reinforced PLA composites

Table 1 presents the effect of the combination between PLA and natural fibre on tensile properties from previous findings. Referring to Table 1, coir, hemp, jute, pineapple leaf fibre (PALF) and ramie are famous PLA reinforcement agents for the study of tensile properties. In addition, most of the combinations between natural fibre and PLA show positive results. Most composite samples present an enhancement in tensile strength compared to virgin PLA. The highest increment was achieved by combining PALF and PLA with a 125% enhancement. Other than that, 30% of fibre percentage shows major optimum fibre content. Meanwhile, the processing method showed a balance between injection moulding and compression moulding, with 11 and 18 uses from previous studies.

The significant increase in tensile strength of PLA composites when the natural fibre is used as a reinforcement agent demonstrates that natural fibre and PLA are compatible. According to a prior study, the excellent wetting of the PLA and the cracking behaviour of the fibre suggested that stress is easily transferred from the matrix to the fibre [62, 63]. This behaviour may be explained by the same hydrophilic properties of natural fibre and PLA [64].

However, the combination of kenaf and coir presents a negative improvement of tensile strength compared to virgin PLA. The combination between kenaf and PLA indicated that tensile strength dropped 40% from the net PLA tensile strength value. Meanwhile, all three findings from previous research regarding coir-PLA composites present -4%, -21% and -58% enhancement in tensile strength results.

Table 1. The previous finding of natural fibre reinforced PLA composites.

Natural fibre	Percentage of fibre (%)	Tensile strength (MPa)		Enhancement (%)	Reference
		Virgin PLA	PLA Composites		
Bamboo	51	60	80	33	[39]
Coir	7	71	56	-21	[40]
Coir	20	56	54	-4	[41]
Coir	30	24	10	-58	[42]
Flax	30	50	53	6	[43]
Hardwood	40	60	100	67	[44]
Hemp	6	51	53	4	[45]
Hemp	30	54	65	20	[46]
Hemp	30	40	45	13	[47]
Hemp	30	5	8	60	[48]
Hemp	40	35	45	27	[49]
Jute	20	52	54	4	[50]
Jute	20	52	55	6	[51]
Jute	30	45	48	7	[52]
Jute	50	50	79	58	[53]
Kenaf	30	58	35	-40	[54]
PALF	10	-	35	-	[55]
PALF	50	8	18	125	[56]
Ramie	30	55	70	27	[57]
Ramie	30	45	52	16	[52]
Ramie	30	17	26	58	[58]
Ramie	30	45	59	31	[59]
Softwood	40	60	95	58	[44]
Sugarcane bagasse	30	55	57	4	[60]
Sugar palm	30	8	14	69	[61]

Ibrahim et. al. mention in their research that the primary factor contributing to the negative impact of kenaf-PLA tensile strength result is weaknesses in the composites fabrication process [54]. The composites' porosity due to fibre agglomerations issue acted as a stress concentrator for crack propagation. A similar case was found in coir-PLA composites that had a negative impact on tensile properties results. The fibre agglomeration and the poor fibre wetting resulted in weak interfacial adhesion between coir fibre and PLA [41, 42]. In addition, Adeniyi et. al. also reported in their review article that untreated coir fibres give poor fibre-matrix adhesion [65].

Additionally, past research has suggested that coir fibre has a low tensile strength compared to other natural fibres, which has had a detrimental influence on the tensile strength enhancement of PLA composites [54, 41]. Fiber tensile strength is proportional to cellulose concentration, and most natural fibres with a high cellulose content have higher tensile strength. The chemical composition and tensile strength values of natural fibres previously employed as reinforcement in PLA composites are shown in Fig. 5.

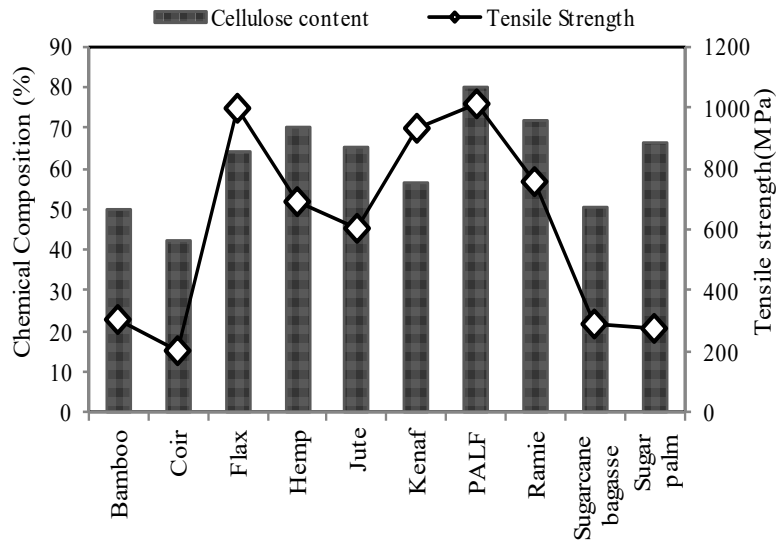


Fig. 5. Comparison of cellulose content and tensile strength of selected natural fibre [66-70].

According to Fig. 5, it has been clearly shown that the PALF is among the highest cellulose content and tensile strength. Meanwhile, coir is contradicted among the lowest cellulose content and thus among the lowest tensile strength. This finding agreed with the above discussion regarding the negative impact of combining coir and PLA. Therefore, selecting suitable natural fibre as reinforcement in PLA composites is one of the essential considerations in manufacturing PLA composites.

3.2. Natural fibre reinforced PHA composites

The study on natural fibre reinforced PHA polymer is still limited, especially in mechanical properties investigation. Table 2 presents the previous studies on using PHA as a matrix in natural fibre composites fabrication. In Table 2, most of the composite samples present negative enhancement in tensile strength compared to virgin PHA, especially for the combination between PHA and bamboo fibre, palm fibre, and recycled cellulose fibre, rice husk and wood fibre. The combination of PHA and rice husk presents the worst effect, with an enhancement of -54%. On the other hand, the previous finding shows that flax-reinforced PHA presents a positive impact. However, the increment of tensile strength only 7 - 8% compared with virgin PHA.

Singh and Mohanty [71] who investigated bamboo reinforced PHBV composites indicate that the poor tensile strength of PHA composites was attributed to the weak interfacial interaction between the fibre and PHBV matrix. In addition, the porosity in the composites structure due to fibre agglomerations issue acted as a stress concentrator for crack propagation [77]. Similar findings were also explained by Persico et. al. in the flexural strength test. The result shows a negative enhancement of -41% for the combination of kenaf and PHBV [78].

Table 2. The previous finding of natural fibre reinforced PHA composites.

Natural fibre	Percentage of fibre (%)	Tensile strength (MPa)		Enhancement (%)	Reference
		Virgin PHA	PHA Composites		
Bamboo fibre - PHBV	40	21.4	17.0	-21	[71]
Flax-PHB	20	30.0	32.0	7	[72]
Flax-PHB	40	38.0	41.0	8	[72]
Palm Fiber - PHA	40	16.7	9.6	-43	[73]
Rice husk - PHA	40	16.2	7.5	-54	[74]
Wood fibre-PHBV	40	21.4	16.8	-21	[75]
Wood - PHBV	50	30.0	27.0	-10	[76]

This drawback can be enhanced by improving the sample preparation method and applying additional processes such as chemical treatment on natural fibre. It is due to the finding from Luo et. al. showing impressive enhancement in tensile strength result of PALF-PHBV composites with 197% [79]. In other hand, Luo et. al. utilized long fibre of PALF as reinforcement with the compression moulding process. In addition, as per discuss earlier, PALF is among the highest cellulose content and tensile strength between all natural fibre. Therefore, this phenomenon shows that natural fibre and PHA combination still have great potential. However, the fibre selection, preparation and manufacturing process play a significant role in producing natural fibre reinforced PHA composites with good mechanical properties.

4. Conclusions

Biopolymers innovations like PLA and PHA provide a solution as an environmentally friendly matrix in manufacturing entirely biodegradable natural fibre composites. The PLA and PHA thermal properties are compatible with natural fibre in the composite's fabrication process. The 160 °C melting point makes PLA and PHA can be made through a biocomposite manufacturing process before the natural fibres are degraded. As a result, the combination of natural fibre and PLA has produced remarkable positive and competitive mechanical properties. On the other hand, PHA also appears as an excellent potential matrix candidate for natural fibre reinforced biopolymer. The present study reveals several vital considerations in PLA and PHA composite fabrication. The selection of natural fibre with high cellulose content and tensile properties is a significant factor in producing better natural fibre reinforced biopolymer composites. Moreover, the fabrication process improvement in avoiding agglomeration and void creation in composites structure also plays a substantial factor in creating good mechanical properties. In conclusion, combining PLA and PHA with natural fibre is a promising candidate for developing entirely renewable and biodegradable composite material.

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