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Impact Property of Flexible Epoxy Treated Natural Fiber Reinforced PLA Composites

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Abstract

The modification is required for more practical applications due to the brittleness of PLA polymer. The improvement of the impact properties of PLA is an addition of fillers or reinforcements. Bamboo fiber, vetiver grass fiber and coconut fiber were used as alternative reinforcements in PLA composites. Injection molded of untreated and flexible epoxy treated composites at various reinforcement content was prepared. The impact strength of natural fiber reinforced PLA composite decreased with the increased of fiber content. The maximum reduction in impact strength was 23.8, 27.3 and 56.2% for bamboo fiber/PLA, vetiver grass fiber/PLA and coconut fiber/PLA composites, respectively. The flexible epoxy surface treatment improved impact property of bamboo fiber/PLA and coconut fiber/PLA composites when compared against the untreated composites. Unlike the other combinations, treated vetiver grass fiber/PLA composite showed less improvement in impact strength when compared with other natural fibers. Bamboo fiber proved to be the most effective reinforcement among all studied reinforcements.

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Keywords: Bamboo fiber; Vetiver grass fiber; Coconut fiber; Flexible epoxy; Polylactic acid

1. Introduction

Fiber reinforced polymer composite materials have played an important role in a variety of applications. As a result of the increasing environmental awareness, the concern for environmental sustainability and the growing global waste problem is increased year by year. Research in the field of polymers and composites from biological sources strives to replace traditional, synthetic ones with ore

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environmentally friendly and sustainable alternatives. Manufacturing of high performance materials from renewable resources is one ambitious aim currently pursued by many researchers. Biocomposite is a material formed by a matrix (resin) and a reinforcement of natural fibers. The developments of biocomposites from biodegradable polymers and natural fibers have attention in the field of composite science [1-2].

A biodegradable polymer is a large molecule composed of repeating subunits, called monomers, which can be broken down by microorganisms. Polylactic acid (PLA) is a biocompatible and biodegradable polymer with a wide rage of applications. PLA has recently attracted a lot of attention from advocates of sustainable development and green chemistry. It is made from renewable resources, and is an environmentally friendly compound with properties that can commercially compete with non-biodegradable polymers such as petroleum-based plastics. The benefits of PLA include low energy to produce and reducing green house gas emission. In addition, crop derived ecologically friendly matrices are gaining ground over the conventional petroleum-based matrices because of environmental problems in disposal as well as concerns over petroleum availability [3-5].

However, as the brittleness problem of PLA, it requires modification for more practical applications. The improvement of the impact properties of PLA is an addition of fillers or reinforcement materials. Natural fiber reinforced polymer composites have been the focus of academic and industrial research interest due to several advantages, such as low cost, high strength-to-weight ratio and recyclability compared to the synthetic fiber composites. The cellulose fibers in natural fiber are aligned along the length of the fiber providing reinforcing effect to the matrix. The quality of the fiber-matrix interface is significant for the application of natural fibers as reinforcement fibers for polymer matrices [6-8]. However, the impact strength is often listed as the major disadvantages of natural fiber reinforced polymer composites in comparison with glass fiber composites. The impact strength of natural fiber reinforced composite can be considerable changed with the alteration of fiber-matrix adhesion by using different types and amounts of coupling agents. The impact strength can be increased by providing flexible interphase regions in the composite or by using impact modifiers. The use of an impact copolymer improves the impact resistance with some reduction in modulus and strength of the composite [9-11]. The flexible interphase between fiber and matrix introduced by coating flexible epoxy resin was also used for improving thermal resistance of natural fiber [12-13]. Moreover, the flexible epoxy resin is considered to be the surface modification for enhancing the impact property of natural fiber reinforced polymer composites.

The purpose of this study was to examine the potential reinforcing effects of short, randomly oriented natural fibers on polylactic acid (PLA). Bamboo fiber, vetiver grass fiber and coconut fibers were chosen as the reinforcement because it is an abundantly natural resource in Thailand. The effect of flexible epoxy resin treated bamboo fiber, vetiver grass fiber and coconut fiber on impact property of composite were investigated.

2. Experimental

2.1. Materials

Polylactic acid (PLA: TE-2000C) with a density of 1.25 g/cm^3 was obtained from Unitika plastics division. The natural fiber consisting of bamboo fiber (BF), vetiver grass fiber (VF) and coconut fiber (CCF) (all materials obtained from Thailand without any pre-sizing treatment) were selected as the reinforcement for PLA. The composition and characteristic of selected natural fibers were listed in Table 1. Flexible epoxy resin (Epoxidized polybutadiene, EPOLEAD PB 3600, Daicel chemical Co.Ltd., Japan) was used as surface treatment for the natural fibers.

Table 1. Physical, mechanical properties and composition of selected natural fibers [3-5]

Natural fiber	Density (g/cm^3)	Modulus (GPa)	Strength (MPa)	Microfibril angle (deg)	Cellulose (%)	Lignin (%)	Average fiber aspect ratio
Bamboo fiber (BF)	0.80	35.9	441	2-10	60.8	32.2	9.5
Vetiver grass fiber (VF)	1.50	12.0-49.8	247-723	N/A	72.6	17.0	3.8
Coconut fiber (CCF)	1.10	4.0-6.0	131-175	30-49	43.0	45.0	26.0

2.2. Specimen preparation

The flexible epoxy resin (1 wt.% of reinforcement) was dissolved in acetone (1 g resin : 200 ml acetone) for reducing the viscosity prior the surface treatment process. The treated natural fibers were first dried at room temperature for 24 hours. Then the treated natural fibers were second dried in vacuum oven at 80^{o}C for 24 hours. The content of natural fibers were varied from 10, 20, 30 and 40 % by weight, respectively. The untreated and treated natural fibers were compounded with PLA matrix by twin screw extruder (JSW TEX30HSS) at 200^{o}C. The dumbbell-shaped testing specimens of untreated and treated natural fibers were fabricated by injection molding (TOYO TI-30F6) at an injection temperature 200^{o}C.

2.3. Testing

Izod impact test was performed on the Digital Impact tester (Toyoseki) with 5.5 J pendulum in accordance with ASTM D256. The dimension of specimen was 10x60 mmxmm. The V-notch shape with 2 mm deep was prepared. At least five specimens were repeated.

2.4. Scanning electron microscopy (SEM)

Scanning electron microscope (JEOL JSM 5200) was conducted on the fracture surface of the tested specimens to examine the failure surface and failure behavior. Specimens were mounted on aluminium holders and gold sputtered for 6 minutes prior observation.

3. Results and discussion

The izod impact strength of composites at different natural fiber content was shown in Fig. 1. The impact strength of pure PLA was 2.05 kJ/m^2. The impact strength of natural fiber reinforced PLA composites decreased with the increased of natural fiber content. This indicated that the addition of natural fiber was ineffective to improve the brittleness of PLA.

The reduction of impact strength of composites was resulted from the poor interfacial adhesion between fiber and matrix as shown in SEM micrograph in Fig. 2a-2c. In addition, there are the differences in impact behavior of different type of fiber of natural fiber reinforced PLA composites. The maximum reduction in impact strength were 23.8, 27.3 and 56.2 % for BF/PLA, VF/PLA and CCF/PLA composites, respectively. It is known that the interfacial adhesion strength between matrix and fiber affected the impact property of composites. Impact energy is dissipated by debonding, fiber and/or matrix fracture and fiber pull out. This was revealed by SEM micrograph in Fig. 2a-2c which shown that the interfacial bonding between bamboo fiber and PLA matrix was the best among these three natural fibers.

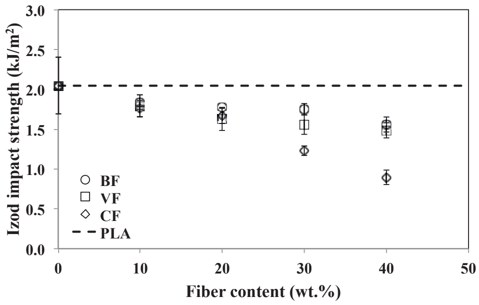

Fig. 1. Impact strength of natural fiber reinforced PLA composite with different fiber content

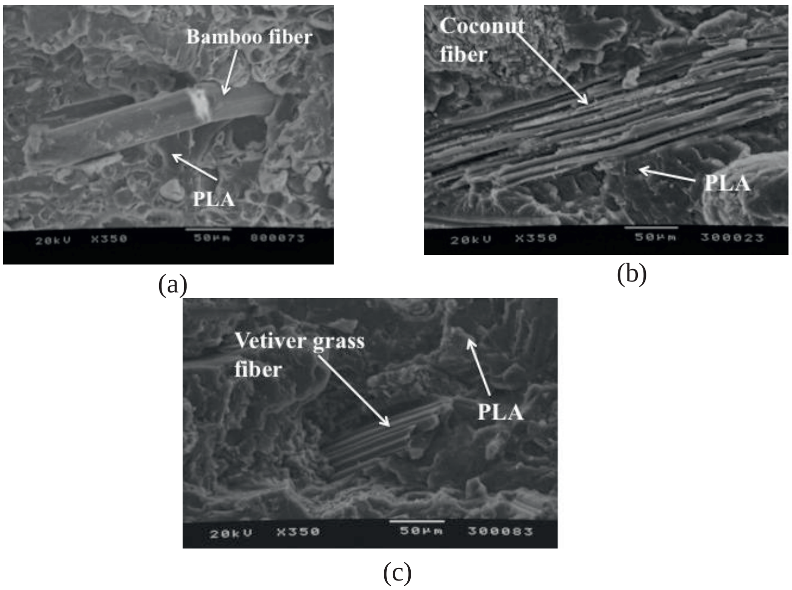

Fig. 2. SEM micrograph of the fracture surface of untreated composites (a) bamboo fiber/PLA;(b) coconut fiber/PLA;(c) vetiver grass fiber/PLA

The impact strength of untreated and flexible epoxy treated bamboo fiber reinforced PLA composites at different fiber content was shown in Fig. 3a. It was found that the impact strength of flexible epoxy treated composites were higher than the untreated composites. In addition, the flexible epoxy treated bamboo fiber at fiber content 40 % by weight showed the impact strength improvement when compared with neat PLA matrix. The flexible epoxy surface treatment also improved the impact property of coconut fiber reinforced PLA composites as shown in Fig. 3b. However, the impact strength of treated coconut fiber reinforced PLA composites was lower than the value of neat PLA matrix. On the other hand, the effect of flexible epoxy surface treatment on the impact strength of vetiver grass fiber reinforced PLA composites was indistinguishable when compared with an untreated composite as observed in Fig. 3c. It can be seen that the effects of flexible epoxy treated on the impact strength improvement were dependent on the type of reinforcement.

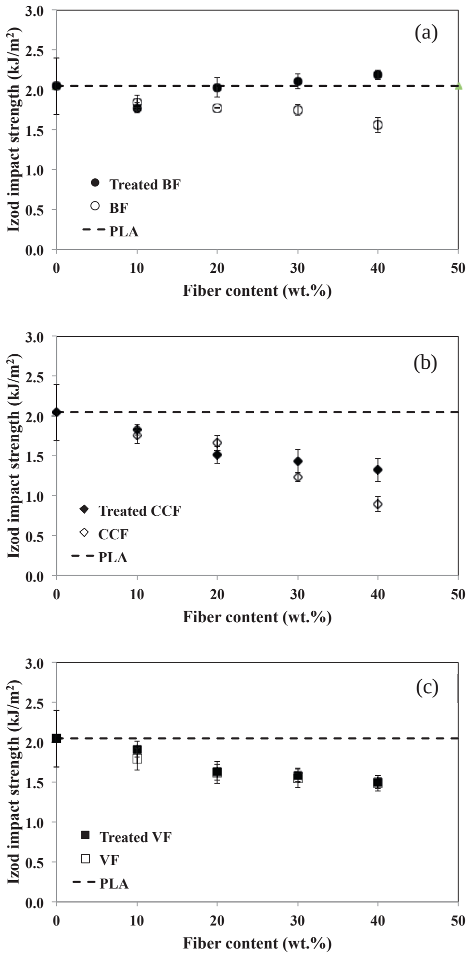

Fig. 3. Impact strength of untreated and flexible epoxy treated PLA composite with different fiber (a) bamboo fiber; (b) coconut fiber; (c) vetiver grass fiber

As the interfacial bonding between fiber and matrix played an important role for the impact strength of fiber reinforced composite. From the SEM micrograph of fracture surface compared between untreated and flexible epoxy treated composites of three different natural fibers in Fig. 4. Through the introduction of flexible interface coupling agent, the likely differences in interfacial adhesion resulted in the different fracture mechanism. As the layer of flexible epoxy resin formed on the surfaces between natural fiber and matrix after treatment as the flexible interphase as illustrated in Fig. 5. The interfacial structure to be formed on fiber surface was not layer but "phase" with thickness was established. The unclear interface between flexible interphase and matrix were formed and called "Thick and Gradient Interphase". This flexible gradient interphase can interrupted the progress of debonding and cracks propagation between fiber/matrix interfaces, which was reported by Yoshikawa et al. [14]. However, in this study the gradient interface between PLA matrix and flexible epoxy interphase was not clearly observed.

The interfacial adhesion between fiber/interphase and interphase/matrix were the important factors for the effectiveness of flexible interphase. It can be seen that at the surface of treated bamboo fiber and coconut fiber in Fig. 4a-4b, small amount of flexible epoxy resin can be observed on the surfaces of the fibers. The good adhesion between reinforcing fiber and flexible epoxy interphase resulted in impact strength improvement of bamboo fiber and coconut fiber reinforced PLA composites. However, in case of treated vetiver grass fiber in Fig. 4c, no flexible epoxy resin appeared on the fiber surfaces. It can be assumed that the interfacial bonding between flexible resin and PLA was constant with different natural fibers. These two different interfacial adhesions resulted in the different fracture mechanism when comparing treated vetiver grass fiber reinforced PLA composite with other reinforcements. It is possible to note that the interfacial adhesion between vetiver grass fiber and flexible interphase was weaker than the interfacial bonding between flexible interphase and PLA matrix. The weak bonding of vetiver grass fiber and flexible epoxy resin resulted in indifferent impact strength improvement versus other natural fibers.

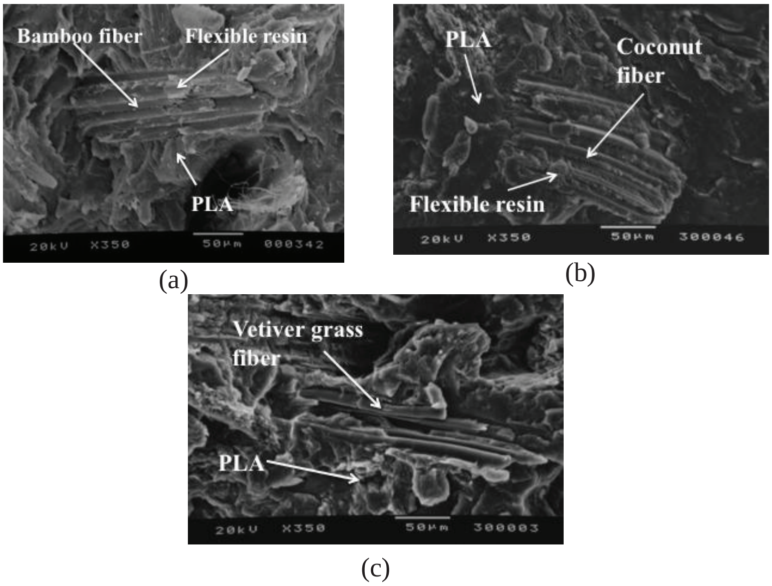

Fig. 4. SEM micrograph of the fracture surface of flexible epoxy treated composites (a) bamboo fiber/PLA;(b) coconut fiber/PLA;(c) vetiver grass fiber/PLA

In Fig. 6 the impact strength improvement performance of untreated composites were compared with flexible epoxy resin treated composites. The comparison was obtained from the differential ratio in percentage of the impact strength at 40 % by weight natural fiber content and in reference to the impact strength of PLA matrix. Among all reinforcement bamboo fiber was the most effective reinforcement to enhance the impact strength of PLA. Moreover, by using the flexible epoxy resin as the surface treatment for bamboo fiber the impact strength of PLA can be improved significantly.

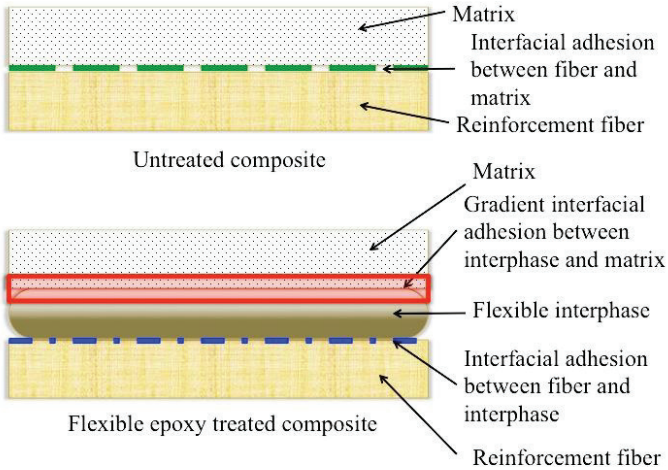

Fig. 5. The schematic illustration of "Thick and Gradient Interphase" concept of flexible interphase treated fiber and matrix

4. Conclusion

The PLA based composite reinforced with different natural fiber consisting of bamboo fiber, vetiver grass fiber and coconut fiber were prepared by injection molding process. The addition of natural fibers reduced the impact strength of PLA composites. The poor interfacial bonding resulted in poor energy dissipation between natural fiber and PLA matrix. The effects of flexible epoxy surface treatment on impact strength of PLA based composites were evaluated. The flexible epoxy surface treatment improved the impact property of bamboo fiber and coconut fiber reinforced PLA composites when compared with untreated composites. However, it can be seen that the flexible epoxy surface treatment was ineffective with the vetiver grass fiber combination. The effectiveness of flexible interphase on mechanical property improvement was not only affected by generated of "Thick and Gradient Interphase" but also the adhesion strength between fiber and the surface treatment. In this study, the bamboo fiber proved to be the most effective reinforcement for the impact strength improvement of natural fiber reinforced PLA composite.

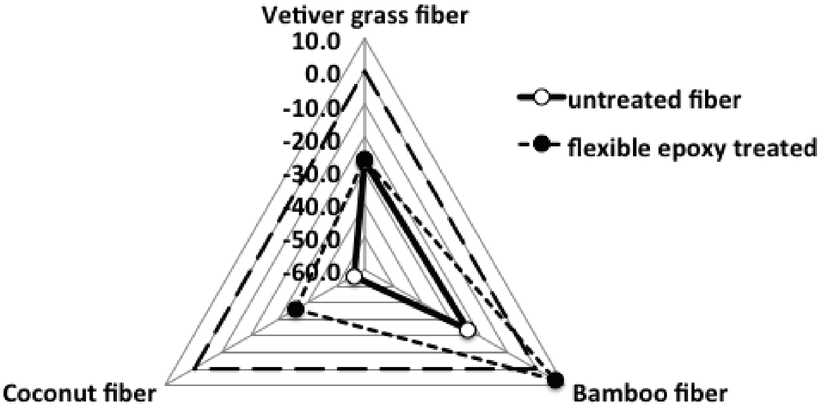

Fig. 6. Impact strength improvement performance comparison between untreated and flexible epoxy treated composites

References

[1] A.K. Mohanty, M. Misra, L.T. Drzal. Sustainable Bio-Composites from Renewable Resources: Opportunities and Challenges in the Green Materials World. *J Polym Envi* 2002;**10**:19-26.

[2] Koichi Goda, Yong Cao. Research and Development of Fully Green Composites Reinforced with Natural Fibers. *J Sol Mech Mater Eng* 2007;**1**:1073-84.

[3] Wendy Ajmass, Allan Amass, Brian Tighe. A Review of Biodegradable Polymers: Uses, Current Developments in the Synthesis and Characterization of Biodegradable Polyesters, Blends of Biodegradable Polymers and Recent Advances in Biodegradation Studies. *Polym Inter* 1998;**47**:89-144.

[4] A.K. Mohanty, M. Misra, L.T. Drzal. *Natural Fibers, Biopolymers, and Biocomposites*. CRC Press; 2005.

[5] Kim L.Pickering. *Properties and performance of natural-fibre composites*. Woodhead Publishing & CRC Press; 2008.

[6] Roger M.Rowell, Raymond A.Young, Judith Rowell. *Paper and Composites from Agro-based Resources*. CRC Press; 1996.

[7] A.K. Bledzki, J. Gassan. Composites reinforced with cellulose based fibres. *Prog Polym* 1999;**24**:221-74.

[8] D.Nabi Saheb, J.P. Jog. Natural fiber polymer composites: A Review. *Adv Polym Technol* 1999;**18**:351-63.

[9] L. Weiloch, D.F. Caulfield, A.R. Sanadi. Toughness improvement in natural fiber reinforced polypropylene. *Internal Report, University of Wisconsin-Madison and Forest Products Laboratoy* 1994.

[10] A. Arbelaiz, B. Fernandez, J.A. Ramos, A. Retegi, R. Llano-Ponte, I Mondragon. Mechanical properties of short flax fibre bundle/polypropylene composites: Influence of matrix/fibre modification, fibre content, water uptake and recycling. *Compos Sci Technol* 2005;**65**:1582-92.

[11] Yupaporn Ruksakulpiwat, Jatuporn Sridee, Nitinat Suppakarn, Wimonlak Sutapun. Improvement of impact property of natural fiber-polypropylene composite by using natural rubber and EPDM rubber. *Compos Part B* 2009;**40**:619-22.

[12] S. Thitithanasarn, K. Yamada, U. Ishiaku, H. Hamada. The Effect of Curative Concentration on Thermal and Mechanical Properties of Flexible Epoxy Coated Jute Fabric Reinforced Polyamide 6 Composites. *O J of Compo Mater* 2012;**2**:133-38.

[13] S. Thitithanasarn, K. Yamada, U.S. Ishiaku, H. Hamada. Jute fabric reinforced engineering thermoplastic sandwich composites I: The effect of molding time. *J Appl Polym Sci* 2013;**127**:2952-59.

[14] Asami Nakai, Akio Ohtani, Takeru Ohki, Toshiko Osada, Masaharu Iwamoto, Hiroyuki Hamada, Nobuo Takeda. Mechanical Properties of FlatBraided Composite with Flexible Interphase. *Proceeding of 18th International Conference on Composite Materials* 2011.