MORPHOLOGICAL AND MECHANICAL PROPERTIES OF A BIOBASED COMPOSITE FROM A LACTIC ACID BASED THERMOSET RESIN AND VISCOSE FIBER REINFORCEMENT

Fatimat .O. Bakare, Dan Åkesson, Mikael Skrifvars, Nima Esmaeili, Shahrzad Javanshir Afshar
School of Engineering, University of Boras
Yanfei Wang
University of South China, China

Abstract

In this study, regenerated cellulose was used to produce thermoset composites from a novel lactic acid (LA)-based resin. Composites were prepared with different fiber alignments (unidirectional, cross wise and non-woven) combined with different fiber loads through compression molding technique in elevated temperature. The biobased composite was characterized with DMTA, flexural test, tensile test and Charpy impact test to investigate the fiber alignments and fiber loads effect on mechanical properties. The aging in high humidity climate chamber was also investigated. The results show that composites can be produced with up to 75% fiber load and with an elastic modulus exceeding 15 GPa in the best cases. This study showed that this environmental friendly composite have comparable mechanical properties with commercial petroleum based glass fiber reinforced (GFR) polyester composites.

Introduction

Recent environmental concerns has led to increasing uses of polymer and composite materials obtained from biomass as raw material for the production of products [1-4], which are used in several fields, such as, automotive, aerospace, construction, marine, consumer goods and industrial applications [5]. In the automotive industry, the main improvement in fuel efficiency is achieved by reducing the mass, which makes manufacturers focus on composite materials because of their low density. Due to government demands on emission reduction and fuel efficiency, the industry are led to produce lighter components from these fiber reinforced plastic composite [6-7].

The production of glass fiber reinforced polyester and epoxy resins leave a large carbon footprint. They are difficult to recycle and dispose of because they do not degrade and linger for years in landfills [8]. This is because they do not degrade and linger for years. So, nowadays researchers focus on composites reinforced with lignocellulosic fibers obtained from renewable sources. These fibers can be classified into two groups, natural (e.g. jute, sisal, bagasse, bast) and man-made fibers (e.g. viscose, rayon). These fibers are advantageous as they are inexpensive, abundant, have low densities and low energy consumption, biodegrading, easy to surface modify, eco-friendly, less abrasive to processing equipment and have high specific strength. Therefore lignocellulosic fibers make them widely acceptable in the automotive and construction industry [9-11].

Much research has been carried out in the production of composites for interior parts of vehicles from natural fiber with polymer matrix because of their low density and cost in order to reduce the use of expensive aramid, glass or carbon fibers. Examples are the development of Mitsubishi Motor's first plant-based resin from polybutylene succinate (PBS) and bamboo fiber

for an interior trim material and also the production of high-durability car floor mat using a polylactic acid (PLA) resin and nylon fibers [12]. Other examples also are the use of epoxy matrix with addition of jute fiber in the manufacturing of door panels in the Mercedes-benz E-class vehicles and the production of spare tires for Raum 2003 model of Toyota vehicle using PLA matrix from sugar cane and sweet potato, reinforced with kenaf fibers [7]. The use of natural fiber since the early 1990s by BMW is another example of increased usage of natural fiber in composite production, which by 2001 the usage of these natural fibers has increased to 4000 tonnes using 80% flax and 20% sisal blend to increase the strength and impact resistance of its composites [13].

For many composite applications, thermosetting resins are preferred since the flow viscosity is lower, thus various impregnation techniques are allowed. Recently, researches have been carried out on the synthesis of thermosetting resins from lactic acid [14-15]. This biobased resin was prepared by direct condensation of pentaerythritol, itaconic acid and lactic acid. The obtained star-shaped molecules with pentaerythritol as the core unit were end-capped by methacrylate groups. This resin had relatively good mechanical properties, but tests also showed that the resin had a relatively high viscosity. Another polyester resin based on lactic acid and glycerol was also produced and it showed better mechanical properties and lower viscosity than for the previous resin with pentaerythritol.

In this study, the polyester resin based on lactic acid and glycerol was combined with viscose fiber reinforcement. Both woven and nonwoven reinforcement shapes were investigated.

Methods

Synthesis

The synthesis was done in two stages:

1. The condensation reaction stage; branched oligomers of glycerol and lactic acid were prepared with chain length n = 3
2. End-functionalization stage; the oligomer was reacted with methacrylic anhydride

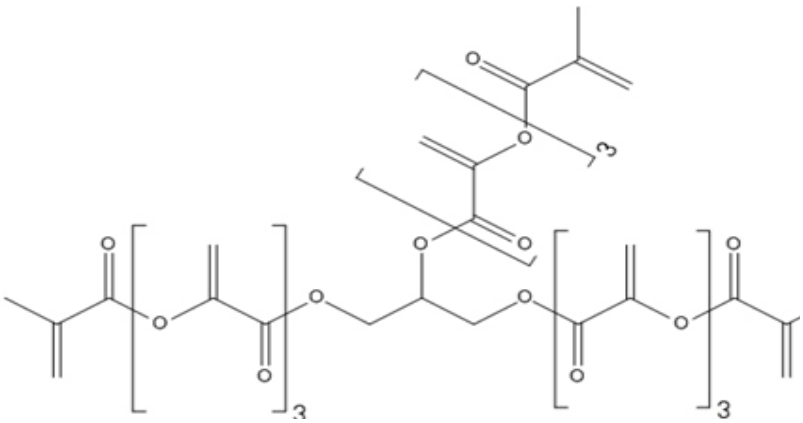

Figure 1: Structure of lactic acid and glycerol (GLA) resin.

Composite Preparation

The reinforcement mats used were viscose non-woven fiber mat with surface weight of 60 g/m^2, and 0.66 mm thickness and was supplied by Suominen Non-Wovens Ltd., Finland. The warp-knitted uniaxial fabric with surface weight of 182 g/m^2 made from a viscose yarn from Cordenka, (twist Z40, linear density of 2440 dtex with 1350 filaments), was delivered by Engtex AB, Sweden. They were cut in 21×21 cm sizes and dried in vacuum oven at 107ºC for two hours, no further pre-treatment was done. The fiber reinforcement was then prepared by a manual layup in three ways: bidirectional (BD) and unidirectional (UD) fiber direction for the woven reinforcement, and the non-woven (NW) fabric. The resin was blended with a thermal initiator and impregnation was done manually by hand spray. Twelve layers of viscose knitted fabrics and 42 layers of non-woven fiber mats were implied for each composite sheet to achieve a thickness between 2 to 3 mm. Different fiber loads were considered (65%, 70% and 75%) for the composite production. The impregnated fabrics were placed in a Rondol hydraulic press to cure at 150˚C for 5 minutes. The composite was characterized using the tensile testing, flexural testing (3 point bending) and Charpy impact testing.

Figure 2: Prepared Viscous- GLA Composite.

Results and Discussion

Tensile testing

The tensile modulus and tensile strength at break for different fiber alignments showed an increase with increasing fiber load from 65 to 75% for UD and BD composites, which can be assigned to a good adhesion between the fiber and the matrix. But for the non-woven the modulus and strength decreases by increasing fiber load from 65 to 75% which can be due to improper impregnation of resin for fiber loads higher than 65%. The results are summarized in Table 1.

Table I: Tensile properties for different composite treatments

Fiber Alignment	Fiber content (wt%)	Tensile Modulus (GPa)	Tensile Strength at break (MPa)
Unidirectional (UD)	65	11.5 (±1.44)	202 (±6.02)
	70	13.1 (±2.69)	214 (±1.72)
	75	14.2 (±1.47)	223 (±6.57)
Bi-directional (BD)	65	7.0 (±0.62)	83 (±12.34)
	70	7.9 (±1.83)	107 (±6.52)
	75	8.5 (±1.09)	114 (±1.72)
Non-woven	65	7.5 (±0.53)	85 (±6.02)
	70	5.5 (±0.55)	63 (±10.40)
	75	5.2 (±1.63)	58 (±29.30)

The standard deviation is given in the brackets.

Flexural testing

The flexural modulus and flexural strength for different fiber alignments increased with increasing fiber load from 65 to 70% for UD and BD composites, but a decrease for fiber load of 75%. But for the non-woven the flexural modulus and strength decreases by increasing fiber load from 65 to 75% which is a sign of improper impregnation for fiber loads higher than 65%. The results are shown in Table 2.

Table 2: Flexural properties for different composite treatments

Fiber Alignment	Fiber content (wt%)	Flexural Modulus (GPa)	Flexural Strength (MPa)
Unidirectional (UD)	65	9.9 (±0.32)	217 (±24.62)
	70	11.6 (±1.21)	228 (±22.19)
	75	11.3 (±0.46)	223 (±13.71)
Bi-directional (BD)	65	5.1 (±0.44)	100 (±2.25)
	70	6.5 (±0.24)	135 (±4.03)
	75	5.9 (±0.31)	114 (±24.13)
Non-woven	65	6.1 (±0.64)	128 (2.66)
	70	5.3 (±0.30)	102 (±7.48)
	75	5.2 (±0.56)	93 (±15.53)

The standard deviation is given in the brackets.

Impact Resistance

The absorbed energy for non-woven composites are much lower compared to aligned fiber composites. Sample investigation showed soft fracture surface and no visible fiber pull out. It could be interpreted that fiber pull out occurred in very short length, which gives lower absorbed dissipation. The results are shown in Table 3.

Table 3: Impact Resistance for different composite treatments

Fiber Alignment	Fiber content (wt%)	Impact Resistance (KJ/m^2)
Unidirectional (UD)	65	132.2 (±30.28)
	70	148.6 (±29.29)
	75	110.9 (±16.47)
Bi-directional (BD)	65	97.2 (±13.58)
	70	113.5 (±12.90)
	75	91.1 (±10.10)
Non-woven	65	14.0 (±2.40)
	70	19.5 (±2.26)
	75	20.0 (±3.96)

The standard deviation is given in the brackets.

Dynamic mechanical thermal testing

The DMTA was done on a Q800 instrument (TA Instruments, DE, USA) and in the dual cantilever bending mode. The specimen dimensions were: length 35 mm, width 8 mm and thickness 2–3 mm. The temperature ramp was from -20 to 150ºC, a heating rate of 3ºC/min, amplitude of 15 µm and frequency of 1 Hz was used. The storage modulus of different composite treatments showed considerable improvement in stiffness of composites compared to neat resin. The tan delta for composites from the neat resin is above 100˚C in all cases, which shows that it will be good for use in many applications. The increase of tan delta with increasing fiber load can be interpreted as good interaction between fiber and matrix. The results are shown in Figure 3.

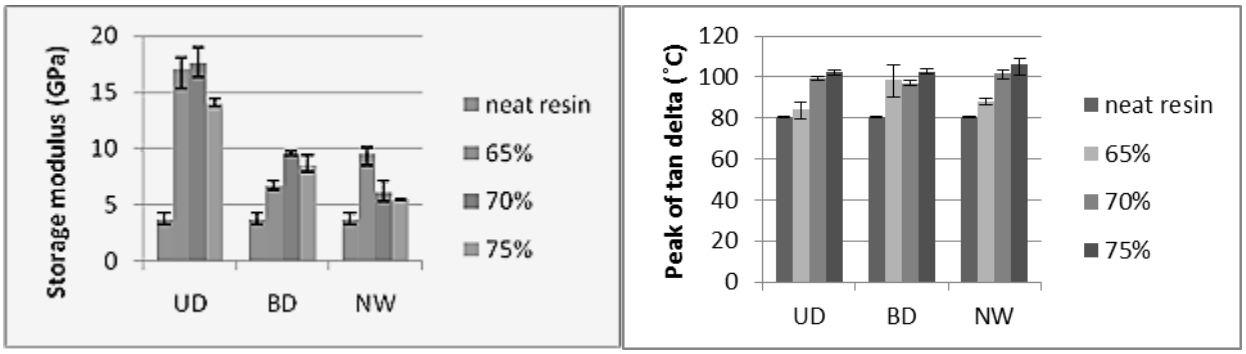

Figure 3: (a) Storage modulus at 25ºC and (b) peak of tan delta of neat resin compared to different composite treatments in room temperature

Ageing test

Table 4 shows the effect on flexural testing and tensile testing for bi-directional composites with 65% fiber load before and after ageing. The results indicates that the mechanical properties of the composite decreases drastically after ageing.

Table 4: Ageing test composites with 65 wt-% bidirectional reinforcement

	Flexural Modulus (GPa)	Flexural Strength (MPa)	Tensile Modulus (GPa)	Tensile Strength at break (MPa)
Before Ageing	5.12	99.65	7.01	83.07
After Ageing	1.92	30.66	2.41	23.22

Summary

A regenerated cellulose fiber from viscose was used to produce a thermoset composite from a polyester resin based on lactic acid and glycerol. The composites were prepared with different fiber alignments (unidirectional, cross wise and non-woven) combined with different fiber loads through compression molding technique at elevated temperature. The results showed that the polyester resin could be used as matrix for cellulosic fibers with fiber load up to 70% and would yield good mechanical properties. Considering its high renewable ratio, low density and relatively low price, cellulosic fibers and lactic acid based resin shows promising

properties for making strong, affordable, bio degradable and renewable resourced composite for many automotive applications.

The ageing experiments showed that during the chosen conditions, ageing deteriorates the composite's mechanical properties, which makes the composites useful mainly for interior applications.

Bibliography

1. Saw, S.K., G. Sarkhel and A. Choudhury, "Preparation and characterization of chemically modified jute-coir hybrid fiber reinforced epoxy novolac composites", Journal of applied polymer science, vol. 125, pp. 3038-3049 (2012).
2. Ramires, E.C., J.D. Megiatto Jr., C. Gardrat, A. Castellan and E. Frollini, "Valorization of an industrial organosolv-sugarcane bagasse lignin: characterization and use as a matrix in biobased composites reinforced with sisal fibers", Biotechnology and Bioengineering, vol. 107, No. 4, pp. 612-621 (2010).
3. Åkesson, D., M. Skrifvars and P. Walkenström, "Preparation of thermoset composites from natural fibers and acrylate modified soybean oil resins", Journal of applied polymer science, vol. 114, pp. 2502-2508 (2009).
4. Ferrero, B., T. Boronat, R. Moriana, O. Fenollar and R. Balart, "Green composites based on wheat gluten matrix and posidonia oceanica waste fibers as reinforcements", Polymer Composites, vol. 34, pp. 1663-1669 (2013).
5. Campanella, A., R.P. Wool, M. Bah, S. Fita and A. Abuobaid, "Composites from northern red oak (Quercus robur) leaves and plant oil-based resins", Journal of applied polymer science, vol. 127, pp. 18-26 (2013).
6. Magurno, A., "Vegetable fibers in automotive interior components", Die Angewandte Makromolekulare Chemie, vol. 272, no. 4751, pp. 99-107 (1999).
7. Koronis, G., A. Silva and M. Fontul, "Green composites: a review of adequate materials for automotive applications", Composites: part b, vol. 44, pp. 120-127 (2013).
8. Marsh, G., "Next step for automotive materials", Materials Today, vol.6, pp. 36-43 (2003).
9. O'Donnell, A., M.A. Dweib and R.P. Wool, "Natural fiber composites with plant oil-based resin", Composites science and technology, vol. 64, pp. 1135-1145 (2004).
10. Ashori, A., "Wood-plastic composites as promising green-composites for automotive industries", Bioresource Technology, vol. 99, pp. 4661-4667 (2008).
11. Oksman, A., M. Skrifvars and J.F. Selin, "Natural fibres as reinforcement in polylactic acid (PLA) composites", Composites science and technology, vol. 63, pp. 1317-1324 (2003).
12. Mitsubishi-motors.com. Mitsubishi Motors develops plant-based green plastic floor mat. Tokyo: Mitsubishi Motors Co. [20.06.06]. <http://www.mitsubishi-motors.com/en/corporate/pressrelease/corporate/detail1475.html>.
13. Jamrichova, Z. and E. Akova, "Mechanical testing of natural fiber composites for automotive industry", University Review, vol. 7, no. 3, pp. 20-25 (2013).
14. Kayode Adekunle, Dan Åkesson, and Mikael Skrifvars, J. Appl. Polym. Sci. 116 (3), 1759 (2010).
15. Dan Åkesson, Mikael Skrifvars, Jukka Seppälä, Minna Turunen, Anna Martinelli, and Aleksandar Matic, J. Appl. Polym. Sci. 115 (1), 480 (2010).