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Investigating the mechanical performance deterioration of Mediterranean cellulosic cypress and pine/polyethylene composites

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Abstract The synergy of the materials physical characteristics, performance and recyclability become vital for industrial sustainability. However, finding a suitable cellulosic fiber type to form potential cellulosic-based composite and investigating performance deteriorations are of paramount importance to expand sustainable design possibilities for various applications. In this work investigations of the mechanical performance deterioration of both Mediterranean cellulosic pine and cypress fibers are experimentally investigated. This was achieved by utilizing the fibers with polyethylene matrix to reveal their potential capabilities for industrial applications. Numerous composites with various parameters like fiber types, fiber loading, fiber size, and reinforcement conditions were designed to study several characteristics of the cellulosic composites, their mechanical performance deteriorations, as well as determining the optimal fiber loading condition for each particular studied mechanical property of the composites. Results demonstrate that mechanical properties are significantly changed with fiber loading. In addition, the failure mode in the high fiber loading composites is an obvious indication of the improper or ineffective load transfer between the matrix and the cellulosic fiber. Moreover, it is

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revealed here that the performance of cypress fibers with polyethylene matrix is much better than that of pine for the considered properties with reference to the neat polyethylene matrix. The overall performance of both types of fibers with polyethylene clearly demonstrates that the performance of cypress fibers is much better than that of pine for all considered properties.

Keywords Bio-composites - Cellulosic fibers - Performance deterioration · Pine · Cypress · Mechanical properties

Introduction

Cellulosic fiber reinforced polymer composites have been utilized and emerged in various industrial applications including structural ones because of their desired properties. This includes the ease of processability, low cost, corrosion resistance, high mechanical properties, high specific strengths and moduli, impact resistance, and high abrasion resistance (AL-Oqla et al. [2014a](#page-6-0), [2016b](#page-7-0); Almagableh et al. [2017](#page-6-0); Goda et al. [2013](#page-7-0); Mansor et al. [2013;](#page-7-0) Methacanon et al. [2010](#page-7-0)). Moreover, high-performance composites are now found in automotive parts, building materials, circuit boards, as well as sporting goods (AL-Oqla and Omari [2017](#page-6-0); AL-Oqla et al. [2014b,](#page-6-0) [2015a](#page-6-0) Shah [2013](#page-7-0)). In addition, the integration among the raw material abilities, cost and performance would significantly

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enhance the industrial sustainability form various economic, environmental and ecological stand points to produce better green products (AL-Oqla et al. [2015b,](#page-6-0) [c](#page-6-0), [e;](#page-7-0) Al-Oqla and Sapuan [2015;](#page-6-0) Haque et al. [2010;](#page-7-0) Ojha et al. [2014](#page-7-0)). However, determining an appropriate cellulosic- based composite type for sustainable industry is still challenging for both designers and the industry, as various parameters are involved.

Materials are practically nominated for a given application based upon their desired properties. From design point of view, engineering components and products are required to bear loads, so the property of chief interest for materials is its strength. However, this strength property alone is not always enough. It is always of paramount importance in modern industries to produce products with light weights (AL-Oqla et al. [2015d,](#page-6-0) [f;](#page-7-0) Sapuan et al. [2016\)](#page-7-0). On the other hand, many applications, like those used in consumer products for casing, exterior decoration, and packaging, are not required to have high mechanical performance that advanced composites possess. Moreover, cheap products that can sustain better environmental performance are strongly recommended to enhance developed more sustainable societies (AL-Oqla et al. [2014c](#page-6-0), [2016a](#page-7-0); AL-Oqla and Sapuan [2014](#page-6-0)). Therefore, natural fiber reinforced polymer composites from available resources are highly recommended as alternative solution for the traditional composites.

Implementing biodegradable and environmentally eco-friendly plant-based lignocellulosic fibers has been admitted as a natural choice for strengthening and filling various types of polymers to make them greener (Aridi et al. [2016b](#page-7-0), [c](#page-7-0); Bajpai et al. [2012](#page-7-0); Thakur et al. [2014b](#page-7-0); Zaman et al. [2012\)](#page-7-0). The availability of inexpensive agro waste cellulosic fibers everywhere in the world has, in its part, fired their use in composite materials due to several advantages like being nonabrasive to processing equipment, easy recycled, $CO₂$ neutral when burned in addition to their good acoustical and thermal properties because of their hollow and cellular nature (Thakur et al. [2014a](#page-7-0)). Their tubular structures moreover reduce their bulk densities and making them lightweight materials. Plant-based fibers such as flax, abaca, hemp, kenaf, bamboo, pineapple, ramie, etc., are being evaluated as low-cost alternative material to reinforce polymers. They are usually obtained from the plant stems or leaves, which are annually renewable comparable to wood. Various characteristics and properties of both natural fibers and synthetic ones are demonstrated in Table [1](#page-2-0) (Khalid Rehman Hakeem et al. [2014](#page-7-0); Thakur [2014\)](#page-7-0).

Several natural fiber composites have been investigated with numerous polymers and fibers for their appropriateness to various industrial applications (Al-Oqla and Omar [2012,](#page-6-0) [2015;](#page-6-0) Aridi et al. [2016a](#page-7-0); Fiore et al. [2016;](#page-7-0) Gupta et al. [2014](#page-7-0)). Such studies involved several mechanical testing, chemical modification for both fillers and polymers as well as proper modeling for selecting the most appropriate composite constituents to maximize the expected beneficial characteristics (Leceta et al. [2014](#page-7-0); Sallih et al. [2014\)](#page-7-0). It is detected that most thermoplastic-based composites utilized for interior and exterior building components are usually made from polyethylene and polypropylene (AlMaadeed et al. [2012](#page-6-0); Kalia et al. [2011;](#page-7-0) Li et al. [2008;](#page-7-0) Sapuan [2013](#page-7-0)). Composites designed from polypropylene and wood flour on the other hand are normally developed for automotive applications and consumer products and recently such composites are being investigated for the use in building applications.

On the other hand, it is obvious that the research and developments in each country are mainly focused on utilizing of locally available fibers. In the country of our interest, Jordan, both pine and cypress trees are dominant sources of fibers that can be utilized for producing natural fiber composites. Moreover, these types of fibers are not frequently investigated for such composites, particularly fibers from Mediterranean countries origin. Therefore, the intention of this work is to experimentally investigate the mechanical performance deterioration of both Pine and Cypress long fibers from Jordan with polyethylene matrix to reveal their potential for industrial applications.

Materials and methods

Materials

Polyethylene resin

Polyethylene (PE) (ExxonMobilTM LLDP LL 8446.21) was utilized to prepare the matrix of the composite as it is a linear low-density butane copolymer designed to have excellent processability, whiteness as well as fast grinding rate. It is a UV stabilized

Fiber				Density $(g cm^{-3})$ Diameter (μ m) Tensile strength (Mpa) Young's modulus (Gpa)	Elongation at break $(\%)$
Flax	1.5	$40 - 600$	345-1500	27.6	$2.7 - 3.2$
Hemp	1.47	$25 - 500$	690	70	1.6
Jute	$1.3 - 1.49$	$25 - 200$	393-800	$13 - 26.5$	$1.16 - 1.5$
Kenaf			930	53	1.6
Ramie	1.55		400-938	$61.4 - 128$	$1.2 - 3.8$
Sisal	1.45	$50 - 200$	468-700	$9.4 - 22$	$3.0 - 7.0$
PALF		$20 - 80$	413-1627	$34.5 - 82.5$	1.6
Abaca		-	430-760	-	-
Oil palm EFB	$0.7 - 1.55$	150-500	248	3.2	25
Cotton	1.5.1.6	$12 - 38$	287-800	$5.5 - 12.6$	$7.0 - 8.0$
Coir	$1.15 - 1.46$	100-460	$131 - 220$	$4 - 6$	$15 - 40$
E-glass	2.55	<17	3400	73	2.5
Kevlar	1.44		3000	60	$2.5 - 3.7$
Carbon	1.78	$5 - 7$	3400a-4800	240b-425	$1.4 - 1.8$

Table 1 Various characteristics of cellulosic and synthetic fibers

resin suitable for excellent dimensional-controlled applications. Its density is 0.936 g/cm³ and its Melt index based on ISO 1133 (190 \degree C/2.16 kg) is 5.0 g/ 10 min whereas its melting temperature is 125 $^{\circ}$ C. The typical mechanical properties of the utilized polymer are shown in Table 2.

Natural fibers

The pine trees are widely available in Jordan. Green leaves of this tree were collected from various regions in Jordan and mixed to make the reinforcing fibers. The length of fibers was between 1.24 and 4.67 cm. These fibers were washed and then dried naturally under the sunlight for 2 days before utilizing in the manufacturing process. Cypress fibers were also collected from various places in Jordan, washed and dried in the same manner of pine ones. The average fiber length was maintained about 5.0 cm. The pine

Table 2 Mechanical properties of the utilized PE resin

and cypress trees as well as their fibers are demonstrated in Fig. [1](#page-3-0).

Samples preparation

The preparation of samples started by weighting 15 g of pine fibers and 100 g of resin, mixed with each other in a random orientation manner of fibers in an iron mould to cover fibers with resin. An insulation paper was used over the mould before mixing for separation. After that the mould was entered into an oven heated up gradually and maintained a 120 $\mathrm{^{\circ}C}$ for 20 min under pressure obtained by a weight over the separated paper. In a similar manner, various samples were prepared with various fiber loadings and fiber types where 15% fiber loading of pine/polyethylene, 15% fiber loading cypress/polyethylene, 30% fiber loading of pine/polyethylene, 30% fiber loading cypress/polyethylene, 45% fiber loading of pine/

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Fig. 1 Cypress and pine trees in addition to their fibers a cypress and b pine

polyethylene, 45% fiber loading cypress/polyethylene, 60% fiber loading pine/polyethylene and 60% fiber loading cypress/polyethylene were prepared to be investigated in addition to the neat polyethylene sample case. The final samples were cut from the prepared plates into a final length of 8 cm as seen in Fig. [2.](#page-4-0) The width and thickness of each sample were measured separately at three different points and the arithmetic average was used in the experimental procedure. The average thickness of the samples was 3.27 mm, whereas the width was 26.46 mm.

Results and discussion

Tensile tests were performed in this work to investigate the mechanical properties of both pine and cypress fibers/polyethylene composites. Two significant properties, which are tensile strength and break strain, were investigated in the tensile tests. The tensile strength property of the produced composites are usually utilized to obtain the maximum force and stress until the composite breaks, which is of paramount importance parameter for the design requirements. Besides, break strain is considered as an important parameter to be investigated in green composites due to its importance for indicating the ductility and amount of energy absorption before breaking. This in order could indicate their appropriateness for various industrial applications like that of automotive applications. After determining the strain gage for each specimen, the universal testing machine was utilized for determining the tensile strength and elongation to break for each sample.

Tests were performed at room temperature and a speed of 100 mm/min. Five trials for each fiber loading sample case were performed for a desired property. The arithmetic average value was then

Fig. 2 Samples prepared for investigation, a a single sample and b different samples in addition to the neat polyethylene one

calculated and utilized for each fiber loading and fiber combination sample. Tests were carried out until specimens break. Various broken samples are illustrated in Fig. 3.

Figure 4 illustrates the influence of the fiber loading parameter on the tensile strength properties of the pine/polyethylene composites. This figure demonstrates that the tensile strengths of the produced composites decrease with increasing the fiber loading.

Fig. 3 Broken samples during tensile tests

Fig. 4 The influence of the fiber loading parameter on the tensile strengths of pine/PE composites

This is mainly due to the poor interfacial bonding between the pine fibers and the polymer. The weak bonding between the hydrophilic fibers and the hydrophobic polyethylene polymer matrix usually obstructs the stress propagation, leading the tensile strength to decrease with increasing fiber loading. Furthermore, poor dispersion usually appears with increasing the fiber loading results in agglomeration of the fibers, ending up with decreasing the tensile properties. It can be detected that 15 wt% of pine fibers/PE composite type has the best performance in resisting stresses with a value of 12.40 MPa, whereas the worst composite type was that of 60 wt% pine fiber.

Figure 5 on the other hand, illustrates the influence of the fiber loading on the break strain properties of the pine/polyethylene composites. It can be revealed here that the elongation to break of the produced composites decrease with increasing the fiber loading. This can be justified as with low fiber loading not much characteristics of the polymer matrix reduced regarding the elongation to break. It is a worthy note here that the elongation to break of the used polyethylene polymer was 145% of the original length. Moreover, it

15wt% Pine/ PE 30wt% Pine/ PE 45wt% Pine/ PE 60wt% Pine/ PE

Fig. 5 The influence of the fiber loading parameter on the break strains of pine/PE composites

can be detected that 15 wt% of pine fibers/PE composite type has the best performance in break strain property with a value of 13.2%, whereas the worst composite type was that of 60 wt% pine fiber with only 9.2%, which implies that a brittle fracture manner occurred in this composite. Generally, results demonstrate that these mechanical properties are significantly changed with fiber loading. Thus, such a failure mode in the high fiber loading composites is an obvious indication of the improper or ineffective load transfer between the matrix and the fiber.

On the other hand, Fig. 6 demonstrates the effect of the fiber loading parameter on the tensile strength properties of the cypress/polyethylene composites. Although similar trends of that of pine fibers appear here, the maximum stress values were more than that of pine/PE composites. In more detail, Fig. 6 also demonstrates that the tensile strengths of the produced composites decrease with increasing the fiber loading. This is again due to the poor interfacial bonding between the pine fibers and the polymer. Furthermore, poor dispersion usually appears with increasing the fiber loading results in agglomeration of the fibers, ending up with decreasing the tensile properties, and this is in agreement with various investigations of cellulosic based composites (Aridi et al. [2016a,](#page-7-0) [c](#page-7-0); Edhirej et al. [2017;](#page-7-0) Essabir et al. [2017\)](#page-7-0), where increasing fiber loading can reduce the composites tensile strengths (AL-Oqla et al. [2017](#page-7-0); AL-Oqla and Omari [2017](#page-6-0); Almagableh et al. [2017;](#page-6-0) Sapuan et al. [2016;](#page-7-0) Symington et al. [2009\)](#page-7-0). However, due to the longer fibers in cypress case, the better performance of the fiber in the composites is noticeable. It can be detected that 15 wt% of cypress fibers/PE composite type has the best performance in resisting stresses with a value of 13.82 MPa, whereas the worst composite type was that of 60 wt% cypress fiber with only 6.25 MPa. Comparable with pine fiber cases, the 60 wt% of cypress causes more agglomeration of the fibers, because it is longer than that of pines results in more improper adhesion with the matrix and thus a decreasing in the tensile properties occurred.

In addition to that, Fig. 7 shows the influence of the fiber loading on the break strain properties of the cypress/polyethylene composites. It can be revealed here that the elongation to break of the produced composites decrease with increasing the fiber loading. This can also be justified by the same reason of that in pine fiber case. Moreover, it can be noticed that 15 wt% of cypress fibers/PE composite type has the best performance in break strain property with a value of 47%, whereas the worst composite type was that of 60 wt% pine fiber with only 19.4%. This in order demonstrated that cypress fibers are much better than pine fibers in case of elongation to break, which reveals its potential for wider applications particularly the automotive ones. Moreover, results demonstrate that these mechanical properties are significantly changed with fiber loading.

Moreover, comparing the composites with various fiber types and fiber loading is presented in Fig. [8.](#page-6-0) It can be clearly noticed that the best performance of pine/PE composite (15 wt%) is less than that performance regarding the elongation to break of the worst fiber loading of cypress/PE ones (60 wt%). This in ordered demonstrate the superiority of the cypress over the pine fibers for structural applications where elongation to break is required.

The overall performance of both types of fibers with polyethylene regarding tensile strengths and break strain is demonstrated in Fig. [9](#page-6-0). It is obvious that the performance of cypress fibers with polyethylene

11.8

9.5

6.3

13.8

2.0 4.0 6.0 8.0 10.0 12.0 14.0 16.0

Max stress (MPa)

Max stress (MPa)

Fig. 7 The influence of the fiber loading parameter on the break strains of cypress/PE composites

Fig. 8 Comparing various composites regarding elongation to break property with various fiber loadings

Fig. 9 The overall performance of the produced composites

matrix is much better than that of pine for both properties with reference to the neat polyethylene matrix.

Conclusions

Both pine and cypress fibers from Mediterranean countries are successfully investigated for the mechanical performance deterioration with polyethylene matrix for the first time. Both types of fibers were utilized as long fibers to reveal their potential for industrial applications. The influence of the fiber loading parameter on the tensile strength properties of the produced composites was demonstrating that their tensile strengths decrease with increasing the fiber loading. Moreover, the influence of the fiber loading on the break strain properties was found to be with the same trend of that for tensile strengths. It can also be revealed here that the elongation to break of the produced composites decrease with increasing the fiber loading. Moreover, it can be detected that that cypress fibers are much better than pine fibers in case of elongation to break, which reveals its potential for wider applications particularly the automotive ones where this particular property is desired.

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