Determining the mechanical properties of biomaterial-based economic thermoplastic composites reinforced with hemp fibres

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ABSTRACT – REZUMAT

Determining the mechanical properties of biomaterial-based economic thermoplastic composites reinforced with hemp fibres

In this study, continuous reinforcement materials of hemp fibres and matrix polymers (staple polylactic acid (PLA), staple bicomponent PLA) were carded in a conventional carding machine. Various proportions of the hemp fibre reinforcement material, ranging from 20%, 30%, 40%, 50%, and 60% were used in the production of biocomposite structures. We have further studied the effect of several carding passages (up to 3 passages) on the strength performance of both the staple PLA and the staple bicomponent PLA matrix. To obtain composite samples, the carded webs were further consolidated in the hot press machine. Further mechanical performance analysis was carried out in both the cross direction as well as parallel to the machine direction. It was observed that the introduction of reinforcing hemp fibre shows an increase in tensile strength up to the critical fibre loading amount and a decrease in tensile strength after the critical fibre loading amount. The highest tensile strength of 35.84 MPa was obtained for (40HEMP / 60PLA / M / P3), applied to the composite structures in the machine production direction, while the lowest value was 14.70 MPa (60HEMP / 40BPLA / M / P1), respectively.

Keywords: *natural fibres, biomaterials, oriented UD tape composites, lightweight materials, bio-composites, staple polylactic acid (PLA), staple bicomponent PLA, hemp fibres*

Determinarea proprietăților mecanice ale compozitelor termoplastice economice pe bază de biomateriale armate cu fibre de cânepă

În acest studiu, materialele de armare continuă cu fibre de cânepă și matrice polimerică (acid polilactic discontinuu – PLA, PLA bicomponentă discontinuă) au fost prelucrate pe o mașină de cardare convențională. Diverse proporții de material de armare cu fibre de cânepă, variind de la 20%, 30%, 40%, 50% și 60% au fost utilizate în producția de structuri biocompozite. A fost studiată în continuare influența numărului de treceri de cardare (până la 3 treceri) asupra performanței de rezistență atât a PLA discontinuu, cât și a matricei bicomponente discontinue de PLA. Pentru a obține probe compozite, vălurile cardate au fost consolidate în continuare în mașina de presare la cald. O analiză suplimentară a performanței mecanice a fost efectuată atât în direcția transversală, cât și în paralel cu direcția mașinii. S-a observat că introducerea fibrei de cânepă pentru armare arată o creștere a rezistenței la tracțiune până la cantitatea de încărcare critică a fibrei și o scădere a rezistenței la tracțiune după cantitatea de încărcare critică a fibrei. Cea mai mare rezistență la tracțiune de 35,84 MPa a fost obținută pentru (40HEMP / 60PLA / M / P3), aplicată structurilor compozite în direcția de producție a mașinii, în timp ce cele mai mici valori au fost de 14,70 MPa (60HEMP / 40BPLA / M / P1).

Cuvinte-cheie*: fibre naturale, biomateriale, compozite cu bandă UD orientată, materiale ușoare, biocompozite, acid polilactic discontinuu (PLA), PLA bicomponentă discontinuă, fibre de cânepă*

INTRODUCTION

The development of structural plant fibre composite components started about 80 years ago and there is considerable interest in them nowadays because of the growing environmental and ecological pressures facing industries. Characteristics and properties of biocomposites have evolved, but improvements are still needed for the effective and durable use of plant fibre reinforcement of composites [1]. As compared to pristine metals and polymers, fibre-reinforced composite materials provide significant advantages in terms of weight, strength, and hardness. However, despite exhibiting good mechanical properties, these materials are prone to the price fluctuations of crude oil-based materials and the high cost of recycling processes of carbon and glass fibres. On the other hand, bast fibre (densities from 1.2 to 1.5 g/cm^3) based reinforced composites can be an alternative to the glass fibre-based (2.6 g/cm^3) reinforced composites, particularly as a low-density replacement, in non-load bearing and partial load-bearing parts. The absence of such natural fibre-reinforced products in structural load-bearing parts has led researchers to alternative studies [1, 2].

Bast fibres such as flax, hemp, and jute are used as reinforcement materials in fibre-reinforced composites as not only do they have long lengths (fibre length 50–120 mm) but also have lower weight and cost than their synthetic counterparts [3]. These “carbon positive” fibres are biodegradable, and recyclable as their carbon dioxide (CO_2) consumption is higher than their emissions [4]. For instance, it has been shown that the cultivation of hemp on an acre absorbs about 2.5 tons of atmospheric CO_2 in one planting season. On the other hand, more than 3 tons of CO_2 are emitted with the production of 1 ton of polypropylene [5].

In addition to the positive environmental effects of the natural fibres, they also provide some cushion towards oil price fluctuations and supply uncertainty. In the global market, the share of natural fibre composites is estimated at 12 billion USD by 2030 with an expected growth of 9 % between 2022–2030 [6]. The increase in the demand for biocomposites in the construction and automotive sectors is being driven by the requirements of superior product properties, environmental sensitivity of the end-users and indeed, legal requirements. The lightweight nature and high rigidity/weight ratio further add to the advantages of using biocomposites [7, 8].

Whilst, based on the fibre length, there are several application methods in the literature for the use of bast fibres (hemp, flax, jute, etc.) in the composites as reinforcement materials. Nonetheless, as discussed below, all the methods exhibit certain advantages and disadvantages:

- The extruded composites produced by mixing short wood-based fibres (acting as filler material, fibre length less than 5 mm) with thermoplastics have poor mechanical properties and their use is limited to non-structural applications [2].
- Short natural fibre nonwovens (usually combined with thermoplastic matrix fibres) are preferred in the composite industry owing to their low cost [9]. These discontinuous fibres are often used for a randomly oriented reinforcement (nonwoven) when there is no preferential tension direction. The random orientation of the fibres in the nonwovens results in an extremely low stress and strain performance in composites. Mechanical properties cannot be designed for non-woven surface applications produced by the random orientation of 30–60 mm fibres [4].
- For the long-spun flax and hemp threads, woven and used as reinforcement elements of composite structures, the optimization of the yarn to be used in textile reinforcement is an important criterion. The tenacity and processability of yarns with fewer twists are low. Woven fabrics produced with spun long linen and hemp yarns are used to reinforce composite structures [10, 11].
- The twist applied to increase the strength of the yarns becomes a disadvantage for the composite structures as it prevents the resin from penetrating the yarn [4, 12].
- In the hybrid yarn production method, the central parallel reinforcing fibres, are wrapped with synthetic filament. In these yarns, the tension applied by the spun yarn to the reinforcement fibres causes the core yarn to displace, which reduces its mechanical properties [2].
- For the woven surfaces produced by the 90° intersection of the yarns in the composites, the intersection points of the yarns lead to the formation of air gaps or localized tension. Therefore, woven natural fibre fabric composite applications do not optimally utilize mechanical strength [4].

For the automotive industry, lightweight structures are an indispensable prerequisite for ensuring high-efficiency, energy-saving vehicles and thus promoting a judicious use of important energy resources. The leading automotive manufacturers are therefore expected to reduce the weight and require composites which are low-priced, extremely lightweight, highly efficient, and more recently, biological-based structural solutions [13].

Studies have shown that natural fibres, especially flax and hemp can be substituted for synthetic composite reinforcements in some cases. There are commercial natural fibre composite products in the market. Therefore, the researchers have increased interest in the subject [10, 14]. For example, the work of Akondaa et al. produced flax/polypropylene unidirectional (UD) thermoplastic tapes using their own developed technology. They obtained 60–110% higher flexural modulus and 35–65% higher tensile modulus results when compared to flax/PP yarn composites. They recognized that the UD flax/PP tapes are an important progress in reinforcing the impact of natural fibres in composite applications [15]. There are commercial UD thermoplastic products in the market one of them is Bpreg's EcoRein®. According to a company statement, the EcoRein® UD family offers thermoplastic-based prepreg reinforced aligned flax fibres which provide high performance possible through fibre direction. The products of Bpreg can be applied as interior trims, body panels of electric vehicles, and trucks by replacing/reducing the use of glass fibre or even carbon fibre in the automotive industry [8, 16]. In a different study, Couture et al. investigated the mechanical properties of two different types of UD flax composites. Aligned flax rovings and flax paper layer were reinforcing filling and PLA was used as the matrix. The mechanical test results have shown that specific tensile properties of the flax/PLA and flax-paper/PLA composites were between 217 to 252 $MPa \times cm^3 \times g^{-1}$. The results of the study are close to woven glass fabrics impregnated with epoxy (227–278 $MPa \times cm^3 \times g^{-1}$) so, it is promising for industrial applications [17].

In this study, the hemp tow reinforcing fibres were intermingled with thermoplastic polylactic acid (PLA) and bicomponent PLA matrix fibres. The bicomponent matrix fibres in the web are heat-treated and shaped under pressure. Composite sheets were produced by melting the thermoplastic fibres. The novelty of this approach is that the application of low-priced hemp tow as reinforcing material without yarn can form textile surfaces. Unlike the numerous process steps of the existing production methods which can affect the production capacity and the costs negatively, our proposed process provides an economical route for the production of the oriented fibre semi-finished products.

MATERIALS AND METHODS

Material

In this study, bio-composites are reinforced with hemp fibres. The properties of the hemp fibres are shown in figure 1. Two different types of bio-fibres, PLA and bicomponent PLA fibres, were used as the matrix materials. The properties of the matrix PLA and the bicomponent PLA fibres are shown in figure 2 and table 1, respectively.

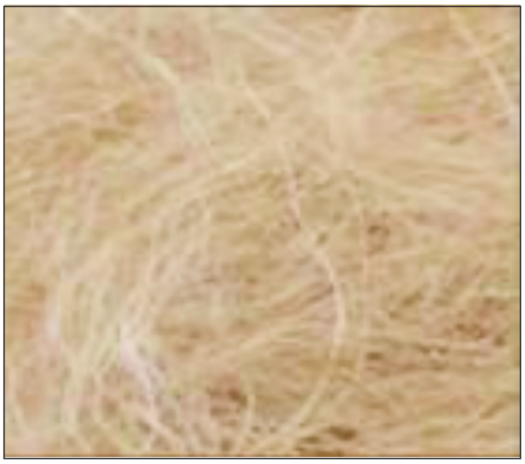

Fig. 1. Image of hemp fibre

Table 1

PLA AND BICOMPONENT PLA PROPERTIES		
Fibre type	**Fibre fineness (denier)**	**Staple length (mm)**
PLA	6.0	51
Bicomponent PLA	4.0	51

The hemp fibre used as reinforcing filler in this study was kindly supplied by Şiteks Şişmanlar Tekstil Sanayi A.Ş., Türkiye. Also, the polymer matrix used is polylactic acid (PLA) and bicomponent polylactic acid (PLA) was provided by Merkas Tekstil Sanayi ve Tic. A.Ş., Türkiye. The bicomponent content of the PLA fibre used a structure with 50% low melting temperature and 50% high melting temperature.

Methods

In this study, a carding process was used to parallelize the staple fibres. Discontinuous bio-fibres of both PLA, biocomponent PLA and hemp fibres were blended in five specified mixing ratios of 20%, 30%, 40%, 50%, and 60% for the production of biocomposite structures. Blended fibres were paralleled to each other and carded in a laboratory carding machine. To determine the effects of the amount of carding passage on fibre orientation, which is one of the main objectives of our study, the webs were produced by passing through three different passes: 1, 2, and 3 passages. A total of 32 samples were produced in line with these variable parameters. The web obtained after combing is wrapped around the drum of the carding machine.

The composite structure was obtained by using the thermoplastic properties of PLA and bicomponent PLA in the hot press machine which is a product of HURSAN and the model number is 50T. The experimental conditions on the hot press were total application time of 2 minutes at 165°C for PLA and 145°C for the bicomponent PLA, respectively. Owing to the heating conditions utilized in the press, the thermoplastic bio fibres melted at the specified temperature and time conditions and contained inside the mould. The composites were further set in the mould with the help of the cooling system present in the press to obtain the final samples. The properties of the produced biocomposite structures are detailed in table 2. The scheme of the application is shown in figure 3. For the tensile strength measurements, the composite plate samples were prepared as specified in the ASTM standard D3039/D3039M-14. These tests were conducted on an INSTRON 4411 testing machine. The samples were selected both in the machine direction, which is the carding direction and in the cross direction. The tensile test was repeated on 20 test samples for each of the settings.

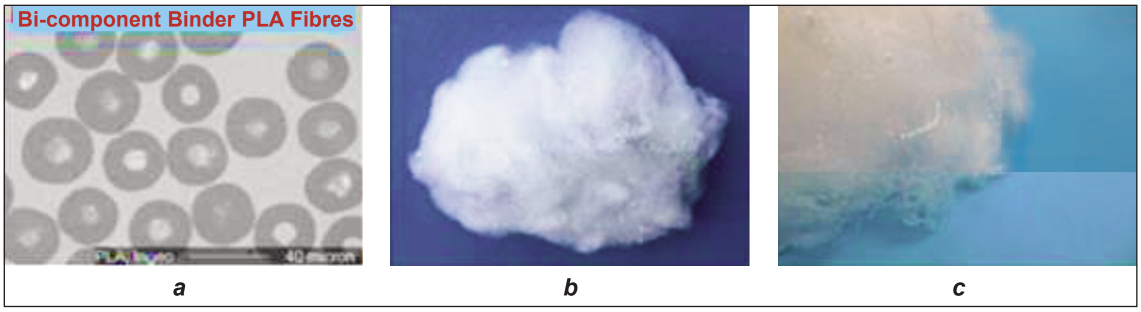

Fig. 2. Images of: *a* – cross-section of Bi-component binder PLA fibres; *b* – PLA fibres; *c* – Bi-component binder PLA fibres

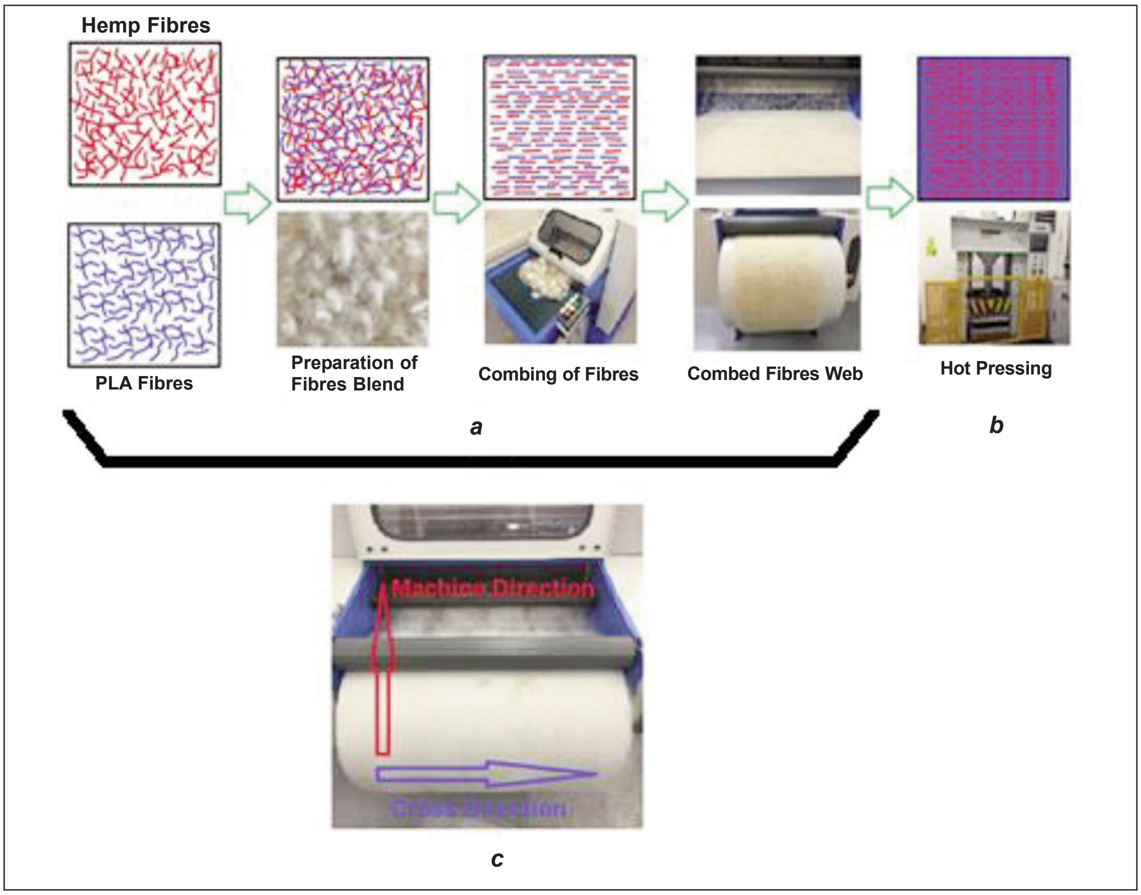

Fig. 3. Schematic drawing of the process: *a* – fibre blend; *b* – pressing process; *c* – machine views of the carding

Table 2

STRUCTURAL PROPERTIES OF BIO-COMPOSITE PLATES

Fibre type	Weight			Thickness			Composition		
	g/m^2	%CV	SS	mm	%CV	SS	HEMP, %wt	PLA, %wt	BPLA, %wt
PLA	275.15	03.34	09.18	0.33	13.64	0.05	-	100	-
BPLA	291.80	09.15	26.71	0.28	11.75	0.03	-	-	100
20HEMP/80BPLA/P1	254.80	05.96	15.20	0.30	13.30	0.04	20	-	80
30HEMP/70BPLA/P1	294.80	09.78	28.82	0.37	18.41	0.07	30	-	70
40HEMP/60BPLA/P1	278.20	08.48	23.59	0.43	16.01	0.07	40	-	60
50HEMP/50BPLA/P1	285.60	09.42	26.89	0.46	14.14	0.07	50	-	50
60HEMP/40BPLA/P1	282.40	11.18	31.56	0.52	14.13	0.07	60	-	40
20HEMP/80BPLA/P2	289.20	08.62	24.93	0.37	15.31	0.06	20	-	80
30HEMP/70BPLA/P2	270.40	08.67	23.45	0.39	13.48	0.05	30	-	70
40HEMP/60BPLA/P2	254.80	11.89	30.30	0.38	13.43	0.05	40	-	60
50HEMP/50BPLA/P2	247.20	11.40	28.18	0.46	12.08	0.06	50	-	50
60HEMP/40BPLA/P2	300.80	09.83	29.55	0.49	11.75	0.06	60	-	40
20HEMP/80BPLA/P3	292.20	08.09	23.63	0.36	15.71	0.06	20	-	80
30HEMP/70BPLA/P3	303.80	10.89	33.07	0.39	13.40	0.05	30	-	70
40HEMP/60BPLA/P3	281.80	09.45	26.64	0.41	14.25	0.06	40	-	60
50HEMP/50BPLA/P3	284.20	10.28	29.21	0.41	13.75	0.06	50	-	50
60HEMP/40BPLA/P3	277.80	10.64	29.55	0.47	13.87	0.06	60	-	40
20HEMP/80PLA/P1	289.46	08.65	25.02	0.25	16.49	0.04	20	80	-
30HEMP/70PLA/P1	293.20	08.89	26.06	0.32	15.56	0.05	30	70	-
40HEMP/60PLA/P1	311.60	08.07	25.16	0.36	17.55	0.06	40	60	-

Table 2 (continuation)

50HEMP/50PLA/P1	298.18	11.85	35.33	0.40	13.61	0.05	50	50	-
60HEMP/40PLA/P1	284.00	09.05	25.69	0.43	10.94	0.05	60	40	-
20HEMP/80PLA/P2	284.60	07.14	20.32	0.24	16.73	0.04	20	80	-
30HEMP/70PLA/P2	300.00	08.35	25.06	0.33	14.83	0.05	30	70	-
40HEMP/60PLA/P2	293.60	07.47	21.92	0.32	15.98	0.05	40	60	-
50HEMP/50PLA/P2	263.60	11.40	30.04	0.34	17.32	0.06	50	50	-
60HEMP/40PLA/P2	289.00	08.34	24.10	0.43	13.82	0.06	60	40	-
20HEMP/80PLA/P3	281.00	09.91	27.86	0.25	15.23	0.04	20	80	-
30HEMP/70PLA/P3	297.00	06.87	20.39	0.25	14.91	0.04	30	70	-
40HEMP/60PLA/P3	272.00	09.01	24.52	0.31	14.72	0.05	40	60	-
50HEMP/50PLA/P3	277.20	09.57	26.54	0.35	14.07	0.05	50	50	-
60HEMP/40PLA/P3	261.80	08.68	22.72	0.39	14.66	0.06	60	40	-

RESULTS AND DISCUSSION

For composite structures consisting of reinforcement and matrix materials, it is known that by increasing the amount of reinforcement material in composites, the strength of the structures can be increased. The mechanical properties of the developed materials mainly depend on the fibre content and their interfacial strength. Generally, the interfacial bonding strength between the hemp fibres and the PLA matrix is weak, which is a disadvantage of the natural fibre composite [18]. However, when the composite reinforcement material reaches saturation, the strength decreases. The reason for this decrease in the strength is caused by exceeding the critical fibre loading amount. In composite structures, the interface between the reinforcement material and the matrix material, as a result of excessive fibre loading, loses strength by not being able to bond with sufficient matrix material [2, 19, 20].

As the fibre web is obtained by carding on a conventional carding machine, the fibres are oriented in cross and machine directions. Consequently, the mechanical strength tests were carried out on the composite surface samples in both these directions.

The results in figure 4 show that for each of the mixture ratios, the strength in the machine direction is higher than the strength in the cross direction. Also, for both PLA and bicomponent PLA, the strength values of the composites increase with the increase of the reinforcement material. The strength value of the PLA matrix is higher than bicomponent PLA, as the PLA matrix creates a better interface with hemp fibre. The difference between these strength values is due to the direction of the fibres in the carding machine. The strength values increase with the

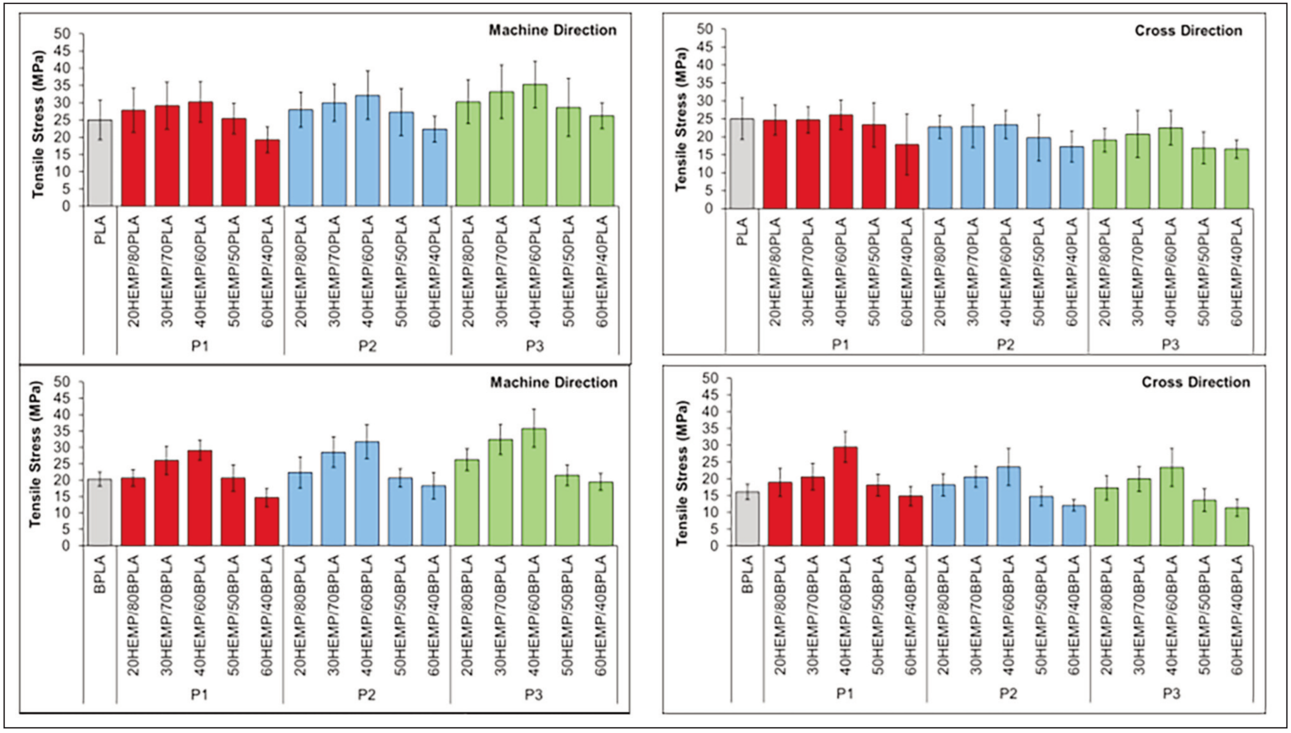

Fig. 4. The effect of reinforcement material on strength in biocomposite structures

increase in the loading amount of the reinforcement material in both the machine direction and the cross direction. The strength increase (by weight) is up to 40% reinforcement material. Since the increase in the reinforcement material after this loading amount causes saturation, the strength decreases above 40% of reinforcement.

The two main components of composite structures are the reinforcing elements and the matrix material. When the fibre web is obtained by the carding process on the carding machine, it is important to control the orientation of the fibres and to have a regular orientation. Multiple passages (three) of the carded hemp fibres were shown to be more effective and enable the control of the orientation of the reinforcement material [2, 19]. The number of carding passages was limited to 3 as it was predicted that a further increase in the number of carding passages would lead to fibre breakage and disorientation.

The results in figure 5 show that the strength in the machine direction is higher than the strength value in the cross direction for each mixture ratio. It is observed that the strength values for both PLA and bicomponent PLA matrices increase in the machine direction with the increase in the number of passages. However, the strength decreases with increasing the number of passages in the cross direction. This is an expected result because the hemp reinforcement material is aligned in the machine production direction.

Looking at figure 6, it can be observed that the elongation amount in the machine direction is higher than the elongation in the cross direction for each mixing ratio. This is because the fibre orientation is in the machine direction in the carding machine [21, 22]. The elongation amounts for both PLA and bicomponent PLA matrices decrease with the increase of the reinforcement material in both the machine direction and the cross direction. The elongation value of the PLA matrix is lower than the bicomponent PLA matrix. This is because the bicomponent PLA matrix partially melts away and the remaining thermoplastic fibres show a higher elongation value than the PLA matrix. In addition, the elongation value of the PLA matrix increased at 20% fibre amount, but with a further increase in the amount of reinforcing fibre, the elongation value decreased as the hemp fibres are of vegetable origin and therefore only allow limited elongation.

As can be observed in figure 7, the elongation value in the machine direction is higher than the elongation value in the cross direction for each mixture ratio. Tensile strength and the modulus of PLA/hemp biocomposites were improved with the increase of adhesion and better composite effect of hemp fibre and PLA. The movement of polymer chains may be restricted by their partial adsorption and by the friction between PLA and hemp fibre, leading to reduced elongation of the obtained biocomposites compared to neat PLA [23]. The elongation amounts of both PLA and bicomponent PLA matrices decrease in the machine direction as well as in the cross direction with the increase of the number of passages applied in the card web production. Since the reinforcement material is plant-based fibre, the elongation amount is limited.

According to the results shown in figure 8, it can be seen that the modulus in the machine direction is higher than the modulus in the cross direction for each of the mixing ratios. The modulus for both PLA

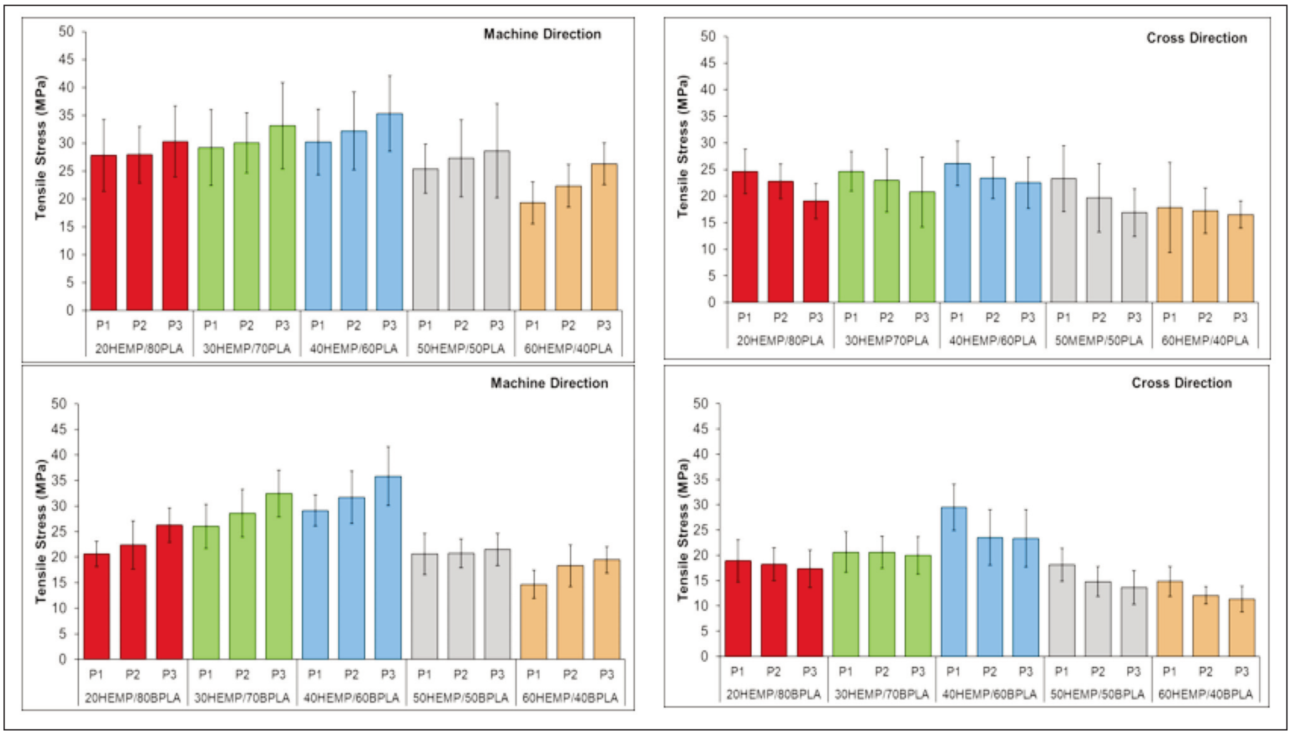

Fig. 5. The effect of the number of carding (passage) of the reinforcement material on the strength of bio-composite structures

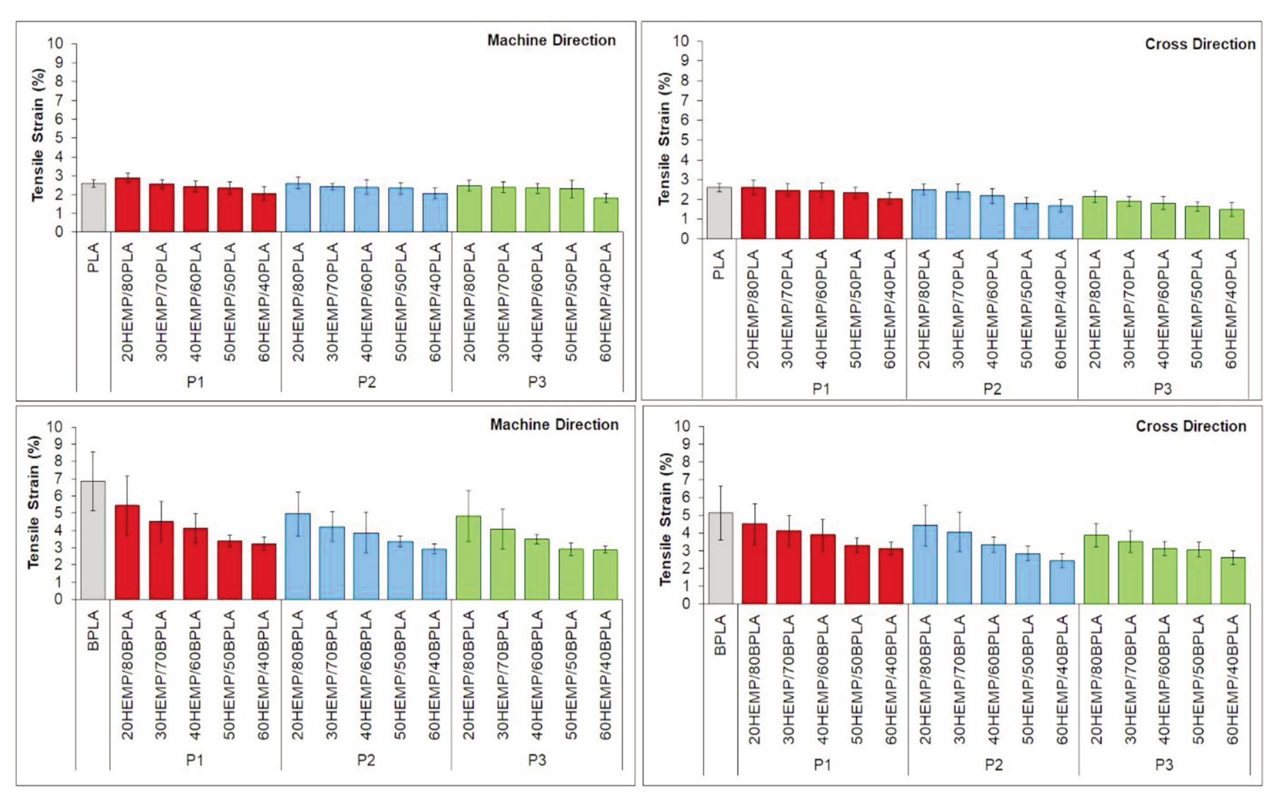

Fig. 6. The effect of reinforcement material on percentage elongation in bio-composite

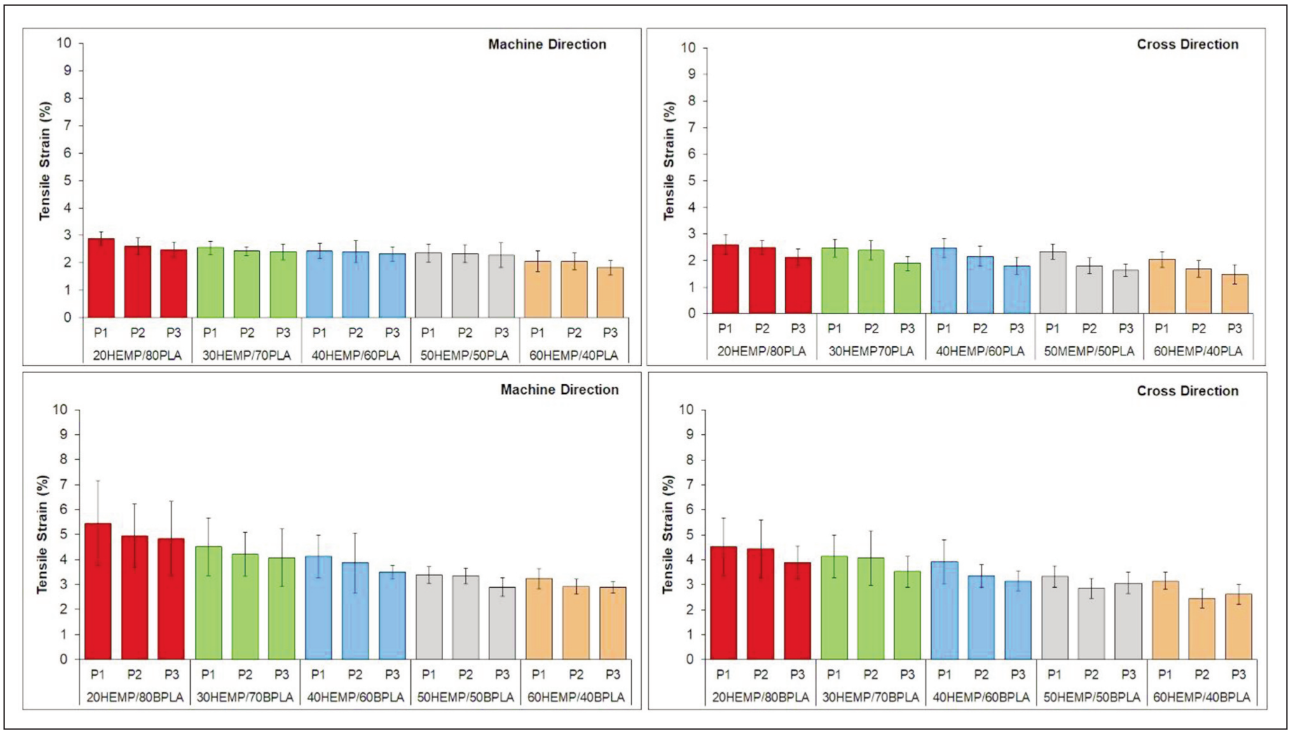

Fig. 7. The effect of the carding passage number of the reinforcement material on the percentage elongation in bio-composite structures

and bicomponent PLA matrices used in the samples increases with the increase of hemp fibre in both the reinforcement machine direction and the direction perpendicular to the machine direction [24]. The modulus value of the PLA matrix in composite samples is higher than the bicomponent PLA as the result of a better interface bonding that has occurred between the PLA matrix and hemp fibre. The reason for the difference between the modulus is the result of orientation in the carding machine. The obtained modulus values increase with the increase in the loading amount of the reinforcement material both in the direction of the reinforcement machine and in the cross direction. The modulus increases for up to 40% reinforcement material and with further loading, reduces.

On examining the results of figure 9, the modulus in the machine direction is higher than the modulus in the cross direction for each mixture ratio. For composite structures produced using PLA and bicomponent PLA matrix materials, the modulus increases in the machine direction as the number of carding web passages increases. However, it is seen that the modulus decreases in the cross direction with the increase of the number of passages applied in the production of the carded web. This is an anticipated

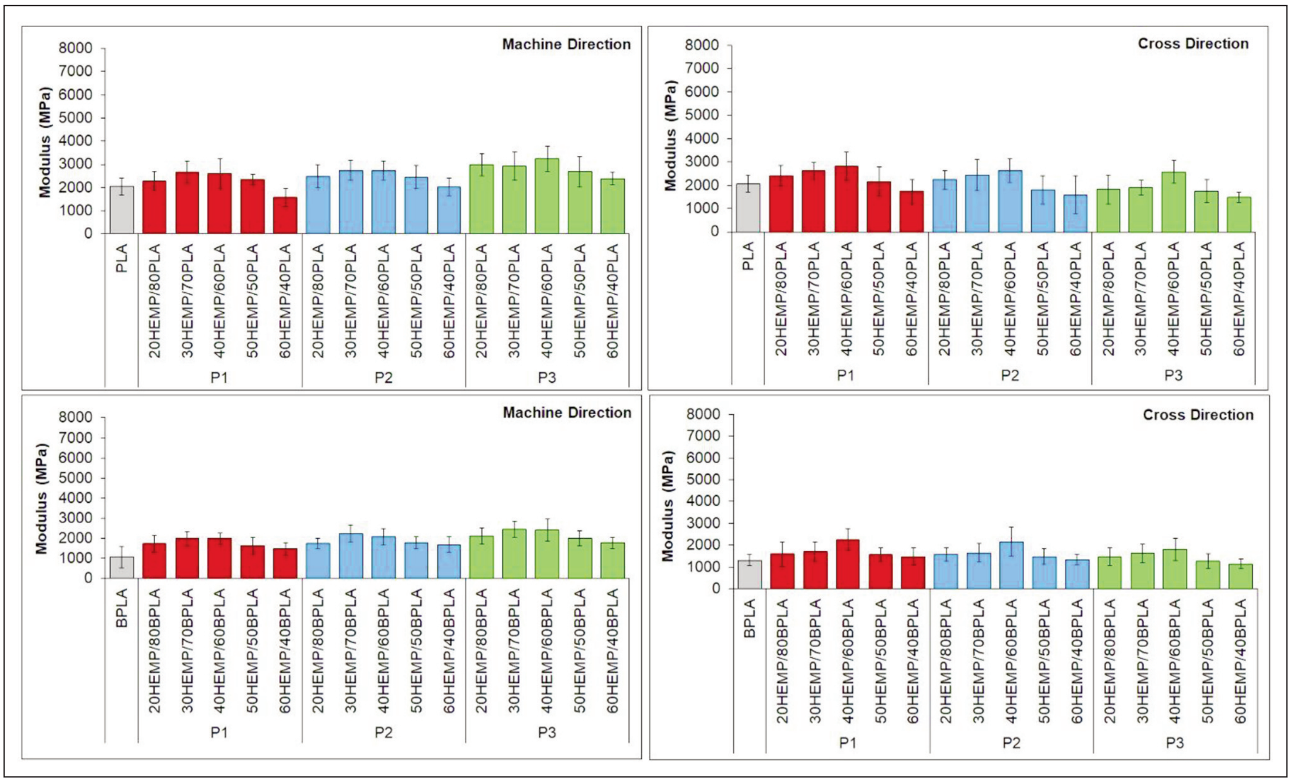

Fig. 8. The effect of reinforcement material on modulus in bio-composite structures

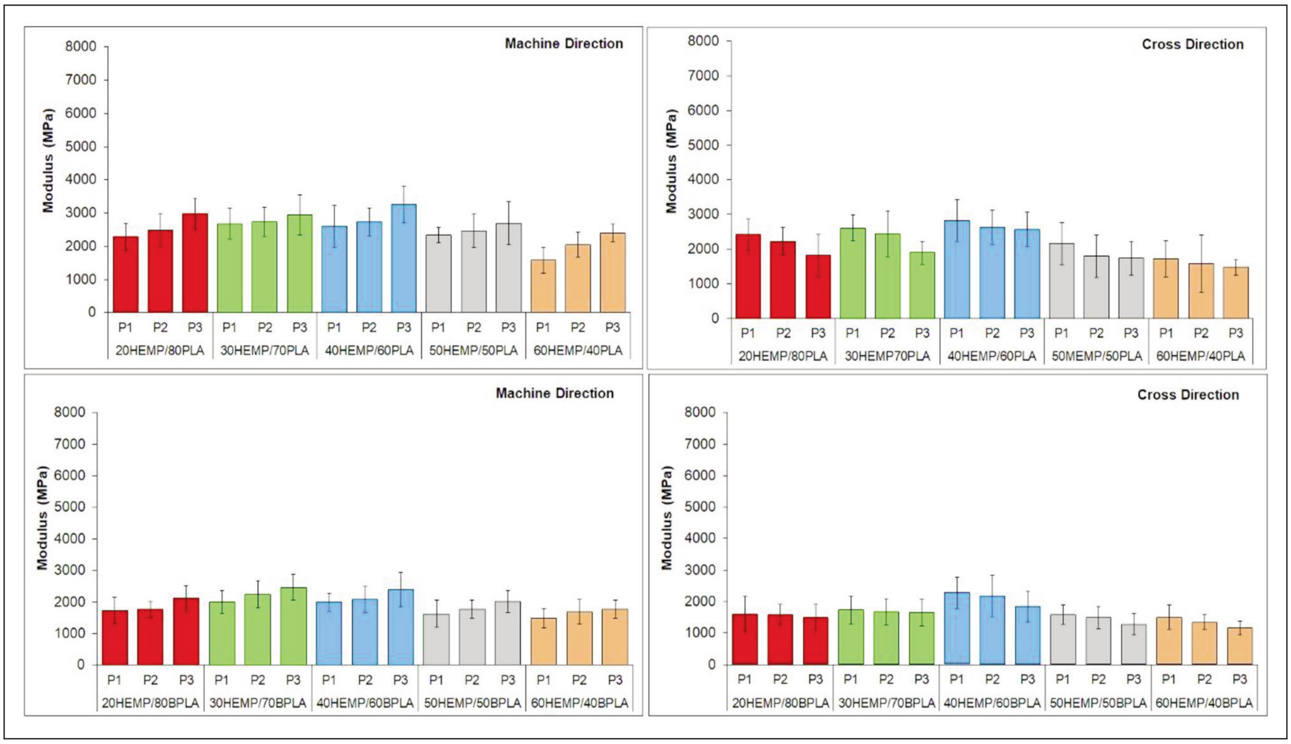

Fig. 9. The effect of the carding passage number of the reinforcement material on the modulus in bio-composite structures

result as the reinforcement material is aligned in the machine direction only [24–26].

CONCLUSION

In this study, bio-based matrix materials of PLA and bicomponent PLA, and natural plant-based hemp fibres were used in the preparation of bio-composites. Matrix and hemp fibres in the continuous form are blended and carded in a conventional carding machine. The carding web was formed in different material amounts ranging from 20%, 30%, 40%, 50%, to 60%. The main aim of the study is to explore the effect of hemp tow proportion, the number of carding passages, and the effect of orientation on the structural properties of fibre-reinforced composite structures. Carding webs containing different amounts of material were consolidated in the hot press process *via* heat and pressure to obtain composite surface samples. Mechanical tests were applied to composite specimens and their structural performances were examined. It was observed that the introduction of reinforcing hemp fibre leads to an increase in the tensile strength up to the critical fibre loading amount with an eventual decrease in the tensile strength beyond the critical fibre loading amount. The highest tensile strength of 35.84 MPa was obtained for the sample (40HEMP / 60PLA / M / P3) in the machine production direction, while the lowest value was 14.70 MPa (60HEMP / 40BPLA / M / P1), respectively. The tensile strength applied to the composite structures perpendicular to the machine direction was highest at 26.13 MPa (40HEMP / 60PLA / M / P3) with the lowest being 11.38 MPa (60HEMP / 40PLA / C / P3), respectively. The tensile strength was shown to increase with an increase in the number of carding passages. The breaking strength obtained in the machine direction is higher than the tensile strength in the cross direction. The strength value of composites obtained from the PLA matrix materials is higher than the bicomponent PLA. The elongation value of the composite structures produced from both the matrix materials decreases both in the machine direction and cross direction with the increase in the number of passages applied in the production of the carding web. The elongation at break in the machine production direction of the composite structures is the highest at 5.47% for 20HEMP / 80BPLA / M / P1 and the lowest at 1.83% (60HEMP / 40PLA / M / P3). The elongation of composite structures' cross direction is the highest at 4.82% (20HEMP / 80BPLA / C / P1) and the lowest at 1.49% (60HEMP / 40PLA / C / P3), respectively. The modulus value of the composites increases with an increase in the loading amount of the reinforcement material (hemp fibre) in both the reinforcement machine direction and the cross direction as well. The internal structures of PLA fibres are different from the internal structures of Bi-component PLA fibres. For this reason, when using PLA as the matrix material, the obtained modulus value Bicomponent PLA is higher. The highest modulus value of the composite structures in the machine production direction is 3245 MPa (40HEMP / 60PLA / M / P3) while the lowest is 1483 MPa (60HEMP / 40BPLA / M / P1). The highest modulus value cross direction of composite structures was obtained as 2823 MPa (40HEMP / 60PLA / C / P1) while the lowest value obtained is 1141 MPa (60HEMP / 40BPLA / C / P3). Hemp tows, a by-product of the hackling process, were used as reinforcement material in this study. The composite structures are just produced with cost-effective carding and hot press processes, without intermediate processes. Lightweight semi-finished composite materials have been produced. The surfaces developed in this study can be used in multilayer lamination applications like the products in the literature and it is thought that they can be used in similar application areas.

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