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Effect of comingling techniques on mechanical properties of natural fibre reinforced cross-ply thermoplastic composites

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Abstract

Continuous natural fibre reinforced thermoplastic composite materials not only offer low weight and better strength than short fibre reinforced composites but are also biodegradable and eco-friendly. The impregnation of resin into the reinforcement is considered as a major concern during the fabrication of thermoplastic composites. Therefore, intermediate materials known as comingled fabrics were developed to assist the fabrication of continuous fibre reinforced thermoplastic composites by aligning the polypropylene fibres alongside the reinforcement natural fibres (jute, hemp and flax) using weaving and knitting techniques. Cross-ply composite panels were fabricated using hot press compression moulding method. The novelty of this work is the simplified methodology to develop the comingled fabrics and the effects of comingling on the mechanical properties of composites. The effect of the comingling technique on the tensile, flexural and impact properties of composites is explained in this research work. Knitted comingled composite specimens exhibit superior mechanical properties than woven comingled composite specimens. The experimental results have shown 14

%, 7 % and 3 % increase in tensile strength, 25 %, 20 % and 13 % increase in flexural strength and 37 %, 54 % and 44 % increase in impact strength of knitted comingled specimens of jute, hemp and flax respectively.

Keywords: A. Polymer matrix composites, B. Mechanical properties, E. Compression moulding, Comingled fabrics

1. Introduction

Natural fibres are gaining remarkable interest in the field of polymer science potentially due to the rising concerns about the environment and global energy crisis [1,2]. The emerging concept of "green composites" have utilized natural fibres in polymer matrix composites to combat the pollution issues and as replacements to the environmentally harmful synthetic materials [3]. Biodegradability makes the natural fibres appealing as the reinforcement in composite materials to reduce waste and landfill dependency. Frequent availability, lightweight, low density, easy to handle, better thermal and acoustic insulation properties, and sustainability are amongst the other key factors making the natural fibres ideal as the reinforcements in polymer matrix composites [4–7]. Natural fibres such as jute, hemp, sisal, kenaf and flax have been identified as suitable reinforcements for thermoplastic composites because of their high mechanical properties [8]. The mechanical properties of natural fibre reinforced thermoplastic composites [9–13] exhibits high specific strength and stiffness at a favourable cost [14–16] for structural applications [17,18]. Thermoplastic composites offer higher fracture toughness and elongation, reduced processing time, the potential for fast, clean and automated manufacturing, and infinite shelf life compared to thermoset composites [19]. The properties of composite often depend on the adhesion between the matrix and reinforcement. Therefore, proper impregnation must be achieved to obtain a reliable composite material for structural applications [20].

The viscosity for most of the thermoplastic matrix are quite high, which inhibits proper impregnation of the matrix to the reinforcement fibres. To overcome this problem, the flow distance during impregnation need to be minimised. Partially impregnated intermediate materials in the form of comingled yarn, thermoplastic film stacking and powder impregnated fibre bundles offer a route for efficient manufacturing of thermoplastic composites due to the reduced flow distance [21].

Comingling is a cost-effective way of blending the reinforcing and matrix constituents. Comingling can be carried out at fibre, yarn or fabric level. Comingled yarns are manufactured by mixing the reinforcing fibres with the thermoplastic matrix fibres using spinning techniques [22] or twisting both the reinforcing and matrix yarns together [23]. Similarly, comingled fabrics can be developed by interlacing or interloping the reinforcing and matrix yarns [24]. Production of structural thermoplastic composites by comingled yarns is one of the most promising routes for composite fabrication as the comingled yarn has a better distribution of matrix and reinforcement in the non-molten state prior to consolidation. Filament winding is commonly used to manufacture preforms of matrix and reinforcing yarns [25,26], but the process is limited to convex shaped components. High mandrel cost, difficulty in changing yarn path in a single layer and unmoulded external surface hindered this approach. Micro braided yarns have also been proposed to overcome the constraint of manufacturing long fibre reinforced thermoplastic composites [27–29], but commercial availability of such systems is restricted at present. In addition, long cycle time and further processes, i.e. preparation of unidirectional or bidirectional preform, are also required. Another approach using handloom unidirectional woven fabrics [19,30,31] have also been proposed, but the fabrication time, yarn tension variation and reproducibility are hindered in the use of the handloom approach. Therefore, faster manufacturing method to tailor the fibre architecture of the preforms is currently in need of development.

Currently, extensive studies on the comingling techniques of continuous yarns are relatively limited. Therefore, in this study, cheap and commercially viable methods are proposed to alleviate the aforementioned problems and the fabric preforms were developed directly by combining the reinforcing and matrix yarns. The novelty of this current work is based on the idea of developing the comingled fabrics on commercially available weaving and knitting machines without any machine or process modifications while keeping the product and process variables constant. Knitted and woven comingled fabrics were manufactured using novel comingling approaches, and then the composite fabrication was carried out using these fabrics. Afterwards, the effect of such comingling techniques on the mechanical properties of cross-ply composites was studied.

2. Experimental detail

2.1. Materials

Hemp and flax 100 % spun yarns were procured from Shanghai Vico Industrial Co. Ltd., China while the jute spun yarn was supplied by Sargodha Jute Mills Limited, Sheikhupura, Pakistan. The hemp and flax yarns were twisted at Nishat Textile Mills Limited, Faisalabad, Pakistan to obtain a constant linear density for all three types of yarns. The characteristics of the reinforcing yarns are described in ***Table 1***. The polypropylene filament yarn (1200 D) was purchased from a local manufacturer based in Lahore, Pakistan.

Table 1

Physical parameters of the reinforcing yarns.

Yarns	Linear density (Ne)	Tenacity (cN/tex)	Elongation (%)	Twist per inch
Jute	2.45 ± 0.15	8.05 ± 0.88	1.62 ± 0.12	7.37 ± 0.25
Hemp	2.45 ± 0.15	13.59 ± 0.98	2.68 ± 0.13	4.95 ± 0.45
Flax	2.45 ± 0.15	18.62 ± 0.90	2.95 ± 0.15	5.68 ± 0.23

2.2. Manufacturing methods

2.2.1. Fabric formation

Comingled fabrics were developed without any treatment using commonly available and broadly used fabric formation techniques, i.e. weaving and knitting. Three different comingled woven fabrics were made using jute, hemp or flax yarn individually in the warp direction and polypropylene yarn in the weft direction (***Fig. 1***) on the Dobby Rapier Loom (BeatMax ISL888-II, Ishikawa Seisakusho Ltd., Japan). 4/1 satin weave with a balanced quantity of twenty ends and picks per inch was selected for the woven fabric since satin weave has less crimp and longer floats which contributes to better mechanical properties [32].

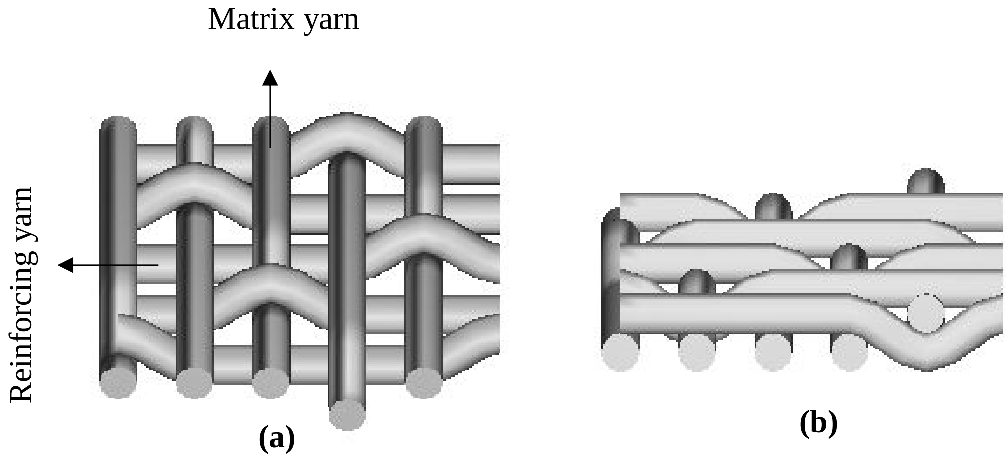


Fig. 1. Schematic representation of comingled woven fabric (a) top view (b) side view.

Comingled knitted fabrics were developed using jute, hemp or flax yarn as the inlaid yarn and the polypropylene yarn as the loop yarn on a fully automatic flat-bed knitting machine (SES 182 FF, Shima Seiki Mfg. Ltd., Japan). The inlaid yarns were trapped by the loops inside the knitted fabrics in a relatively straight configuration (***Fig. 2***). The courses per inch were kept constant (twenty) as compared to the woven fabric for symmetry, while wales per inch were eight. The ratio of reinforcing yarns in both knitted and woven comingled fabrics was the same (47 ± 1 % by weight) which can be ascribed to the similar fineness of all three reinforcing yarns (2.45 ± 0.15 Ne) and the similar number of courses and warps per inch in the knitted and woven fabrics (twenty). The area density of the knitted and woven comingled fabrics was 450 ± 10 g/m^2 measured using the GSM cutter and weighing balance (ASTM D3776).

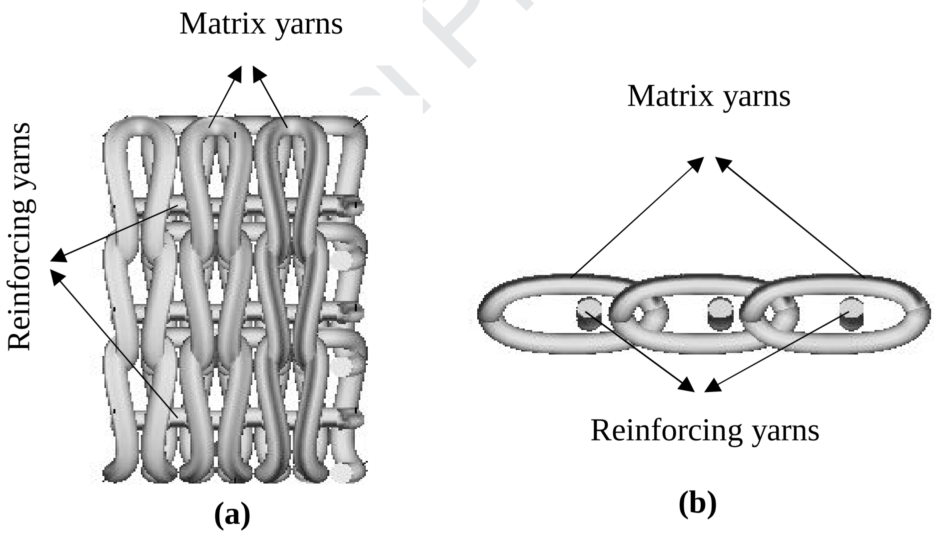


Fig. 2. Schematic representation of comingled knitted fabric (a) top view (b) side view.

2.2.2. Composite fabrication

Cross-ply composites were manufactured using either woven or knitted comingled fabrics exclusively with compression moulding technique. Six layers of fabric were used to manufacture a cross-ply composite $[0°/90°]_3$ at a mould temperature of 185 °C and pressure of 500 Psi for 20 minutes. Cooling was conducted inside the mould for 3 ~ 4 hours under pressure. The fibre volume fraction was 35 ± 2 % as calculated using the rule of mixture using equation (1).

$$V_f = \frac{\frac{M_f}{\rho_f}}{\frac{M_f}{\rho_f} + \frac{M_m}{\rho_m}} \tag{1}$$

where V_f is the fibre volume fraction, M_f is the weight of the fibres, M_m is the weight of the matrix, ρ_f is the density of fibres and ρ_m is the density of the matrix.

2.2.3. Characterization

Natural fibres are prone to thermal degradation due to the presence of lignocellulose. Therefore, it is significant to understand the thermal degradation of natural fibres prior to composite fabrication. Thermogravimetric analysis (TGA) was conducted on a thermogravimetric analyzer, (TG 209 F1 Libra, NETZSCH Group, Germany) by measuring the mass of the sample over time as the temperature was increased to identify the degradation behaviour of the reinforcing fibres. The crucible used for TGA was Al_2O_3, and the temperature range was from 25 °C to 500 °C.

The tensile strength of the natural fibre and matrix yarns was determined by the single-strand method in accordance with ASTM D2256. The tensile tests were conducted on a Single Fibre Strength Tester, (M.250-2.5CT, Testometric Company, UK) at a crosshead speed of 25 mm/min and a gauge length of 250 mm after conditioning the samples at the temperature of 21.4 °C and relative humidity of 64.6 % without pre-tension.

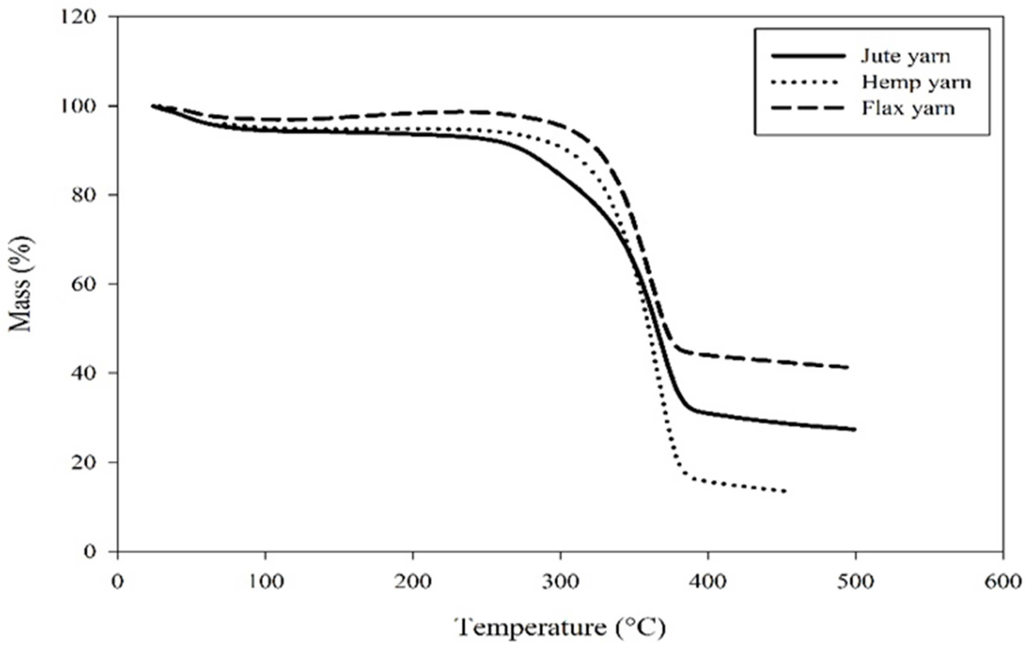
The mechanical properties i.e. the tensile strength (ASTM D3039) and the flexural strength (ASTM D7264) for a total of six composite samples respectively, were measured using a Universal Testing Machine (UTM) (Zwick/Roell, Germany). The

Charpy impact strength (ISO 179) and the drop weight impact strength (ASTM D7136) was measured using an Impact Tester (Zwick/Roell, Germany). The impact strengths of the un-notched samples were measured with Charpy test at a hammer angle of 148° and hammer energy of 50 J while the impact strength by drop weight method was conducted at 5 J energy with a striker diameter of 16 mm.

3. Results and discussion

3.1. Thermogravimetric analysis

The thermal stability of the reinforcing fibres: jute, hemp and flax yarn was evaluated with TGA within the range of 25 °C ~ 500 °C at a heating rate of 10 °C /min. Flax yarn was observed to have better thermal stability than hemp and jute yarn while jute yarn has a higher degradation rate than the other two yarns (***Fig. 3***). This degradation behaviour of the reinforcing yarns can be attributed to the variation of their chemical composition (cellulose, hemicellulose, lignin, pectin, etc.) [33–35]. A three-stage weight loss was observed from the TGA curve. The degradation below 100 °C was due to the evaporation of the absorbed moisture. The second and third stage was



within the temperature range of 230 °C ~ 280 °C and 350 °C ~ 400 °C which indicated the degradation of low molecular weight of the hemicellulose and lignin respectively.

Fig. 3. Thermogravimetric analysis of reinforcing fibres

There was no significant mass reduction observed up to 260 °C. The presence mass reduction would have indicated the degradation of the compositions of the yarns. Therefore, it can be concluded that these reinforcing yarns could undergo composite fabrication below 260 °C without compromising the final product quality.

3.2. Tensile strength

The single-strand method was used to measure the tensile strength of the reinforcing and matrix yarns. It was observed that among the reinforcing materials, flax yarn has the highest tensile strength followed by jute and hemp yarn. Comparatively, the tensile strength of flax yarn was almost equal to the polypropylene matrix yarn. This high strength can be attributed to the better homogeneous nature, higher amount of cellulose, less microfibrillar angle [18,36] and less ligneous residues of the flax yarn [37] (***Table 2***).

Table 2

Chemical composition of the reinforcing fibres [36,38]

Fibres	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Microfibrillar angle
Jute	61-72	13	14	8
Hemp	70	6	22	6
Flax	70-73	2	18-20	5

Growing conditions, harvesting time, extraction method, treatment and storage procedures are among the influential factors other than the chemical composition and the structure of fibres which contributed to the variations in the mechanical properties natural fibres. The tensile failure of the yarns is a combination of fibre slippage and

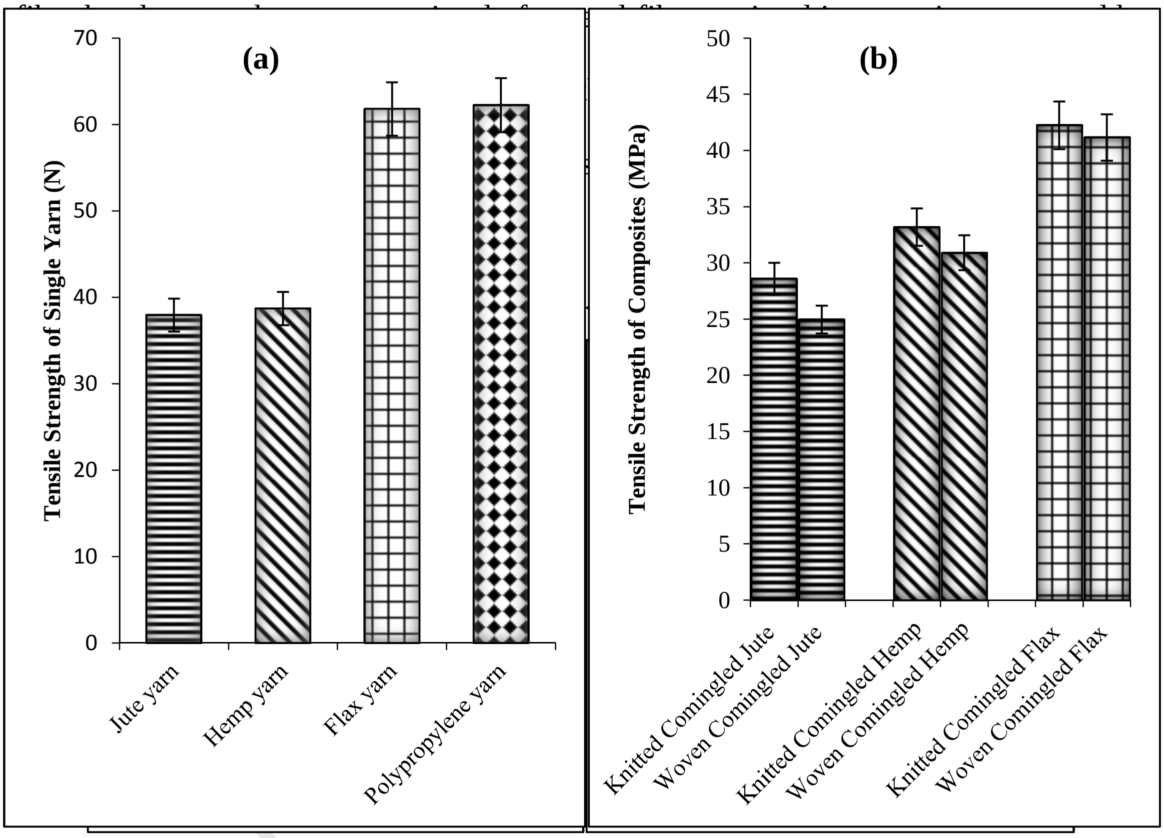
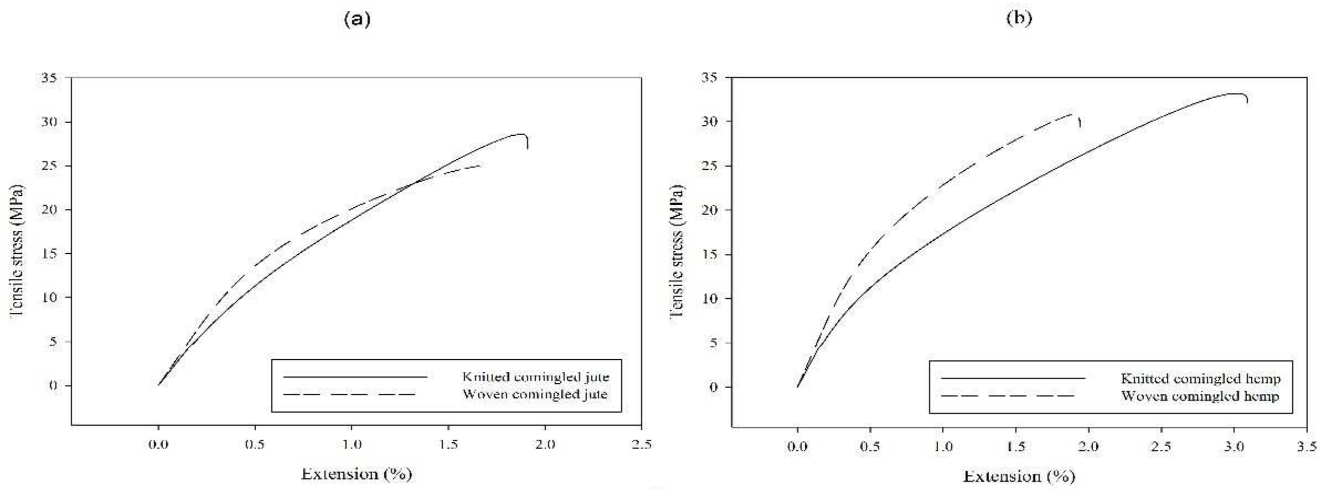
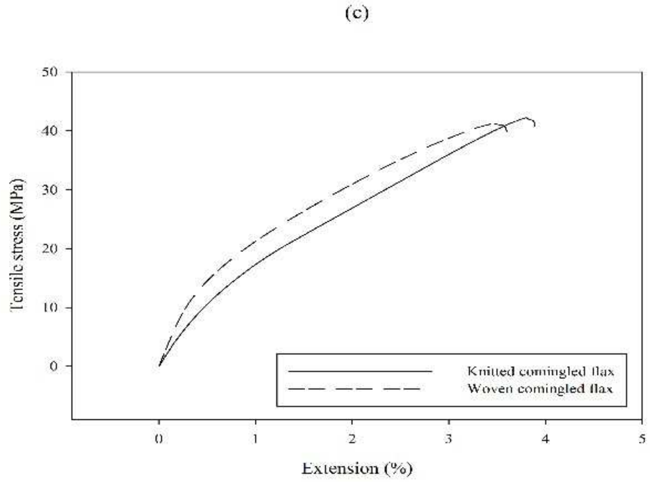


Fig. 4. Tensile strength of (a) single yarns and (b) composites

The composite samples fabricated with flax comingled fabric shown higher tensile strength as compared to others. Knitted comingled samples exhibited higher tensile strength (14 %, 7 %, 3 %) than the woven comingled samples of jute, hemp and flax respectively. Inside the knitted comingled fabric samples, the reinforcing yarn was observed to be entirely covered with matrix yarn from all sides (***Fig. 2***), which





translated into a reduced required flow distance and consequently improved the impregnation between the matrix and reinforcement. However, in the case of the woven comingled samples, the polypropylene yarn was used as the weft yarn, which was interlaced at specific points as per weave pattern (***Fig. 1***). Therefore, the reinforcing yarn was not fully enveloped by the matrix yarn, which then leads to higher required flow distance and lesser impregnation between the matrix and reinforcement.

Fig. 5. Stress strain curves for the tensile test of woven and knitted comingled composites of (a) jute, (b) hemp, (c) flax

Significant increase to the tensile strength was observed in the knitted comingled jute samples compared to the woven samples. It was partly due to a high amount of protruding fibres in jute yarn which hinders in the flow of matrix. The knitted comingling technique reduced the flow distance and hence improved the tensile properties of the jute composites. The stress-strain curves of the composite specimens tested until tensile failure (***Fig. 5***) indicated that all samples have not failed abruptly but rather behaved in a pseudo-plastic manner before attaining the maximum stress value.

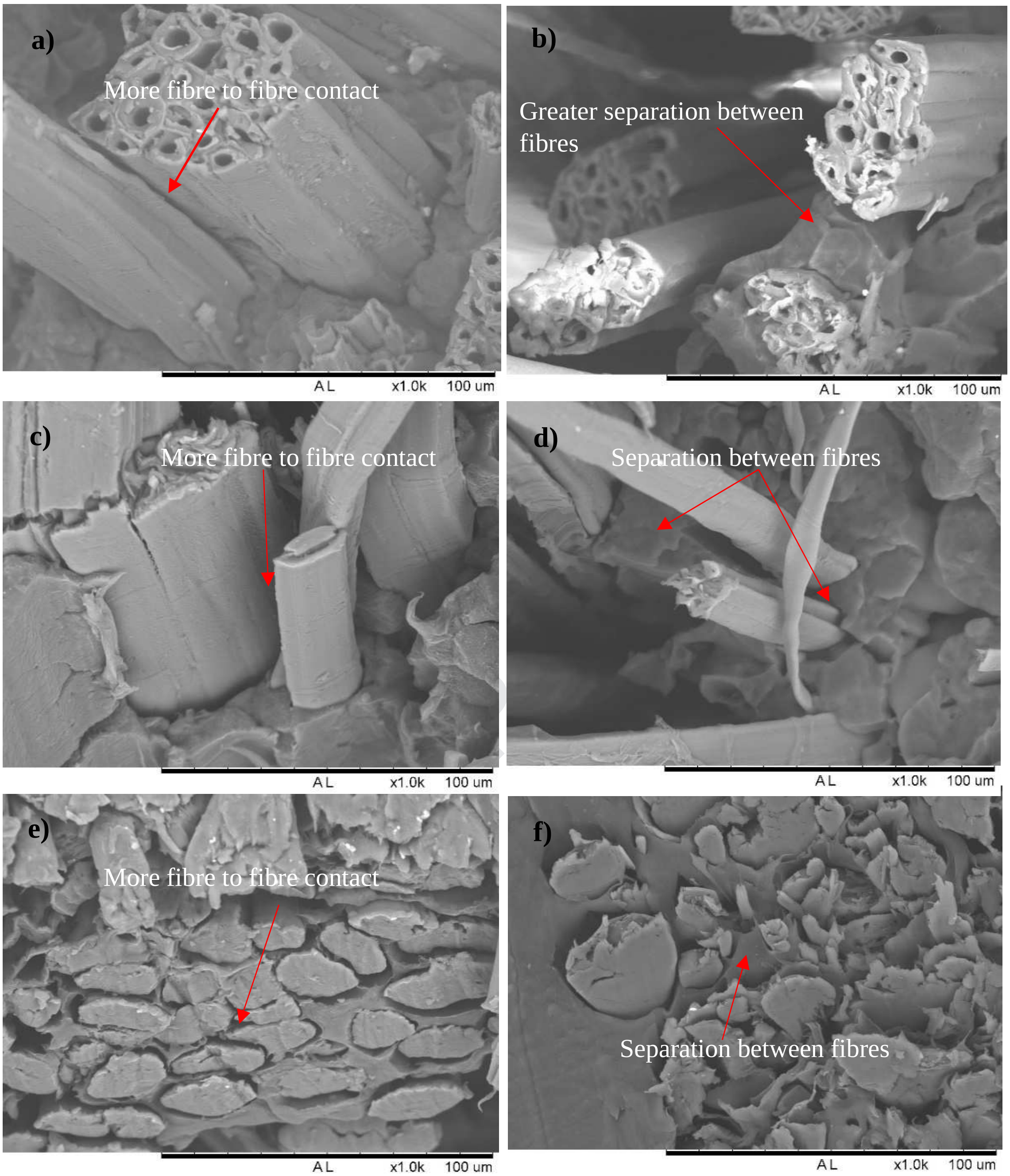


Fig. 6. The SEM images of the impregnation behaviour of the composites (a) woven comingled jute, (b) knitted comingled jute. (c) woven comingled hemp, (d) knitted comingled hemp, (e) woven comingled flax, (f) knitted comingled flax

The separation between the fibres in the SEM images of the composite samples (**Fig. *6***) verifies the differences in the matrix impregnation due to the wetting of the fibres. The fibres of woven comingled composites were tightly packed, which indicates that the matrix has not fully penetrated between the fibres, while in the knitted comingled composites, the matrix has thoroughly wetted the fibres. However, the bonding between the reinforcement and matrix was not good as fibre-matrix debonding was observed in the SEM images.

3.3. Flexural strength

Matrix micro-cracks are the initial damage that occurs under flexural load, particularly on the side of the composites enduring tension. The matrix micro cracks propagate slowly without leading to critical load drop until the development of kink band on the compression side resulted in deformation [39]. Woven and knitted comingled samples of jute, hemp and flax undergo three-point bending test, which yielded the same trend as tensile results (***Fig. 7***).

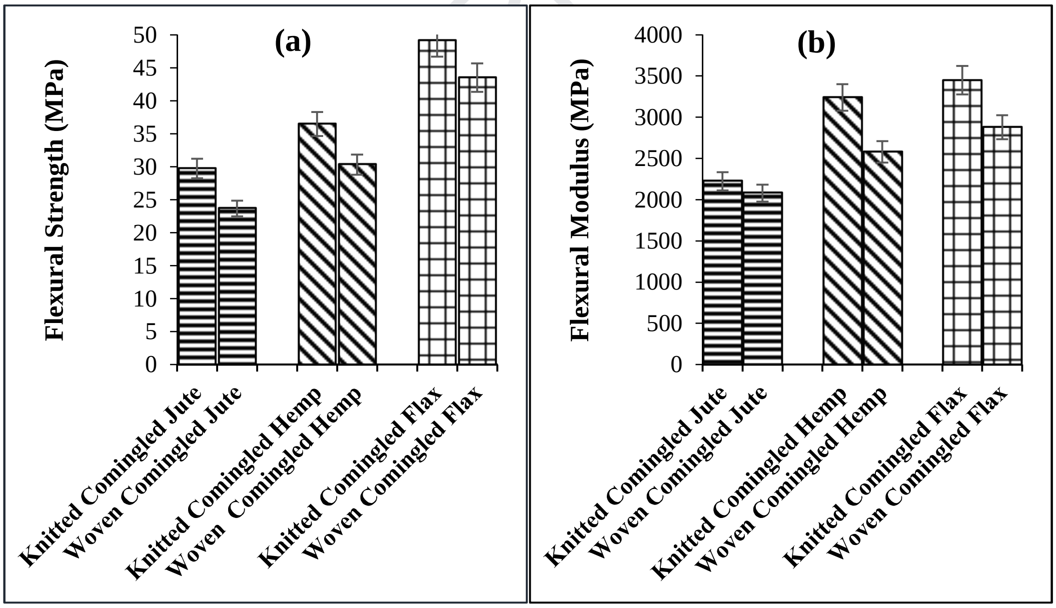


Fig. 7. Flexural behaviour of composites (a) strength and (b) modulus

It was found that the composites fabricated from knitted comingled fabric had considerably higher flexural strength and modulus than composites fabricated by woven comingled fabric. In comparison to the flexural strength of the woven comingled fabric composites, there was an improvement of 26 % for jute, 20 % for hemp and 13 % for flax composites manufactured from knitted comingled fabrics. The effect of the comingling technique on the flexural stress is shown in ***Error! Reference source not found.***. A considerable difference in the flexural behaviour was observed by varying the comingling technique. The inner layers of the laminates was subjected to compressive effect due to the application of flexural loading, while the outer layers were subjected to tensile behaviour [40]. The knitted comingling technique has enabled the polypropylene matrix to fully penetrate between the reinforcement yarns and thus transfers the applied flexural load to the yarns, which then leads to the observed increase in the flexural strength.

In comparison to the flexural modulus of woven comingled fabric composites, the knitted comingled composites had improvement of 7 % (jute composites), 26 % (hemp composites) and 20 % (flax composites). The reduction in flexural strength and modulus of the woven comingled laminates can be attributed to the waviness of the reinforcing yarns. Out-of-plane waviness was observed in the optical microscopic images (***Fig. 9***) for the woven comingled laminates. In general, the published literature [41–43] validated the notion that the increase in fibre waviness can decrease the mechanical properties of the composites.

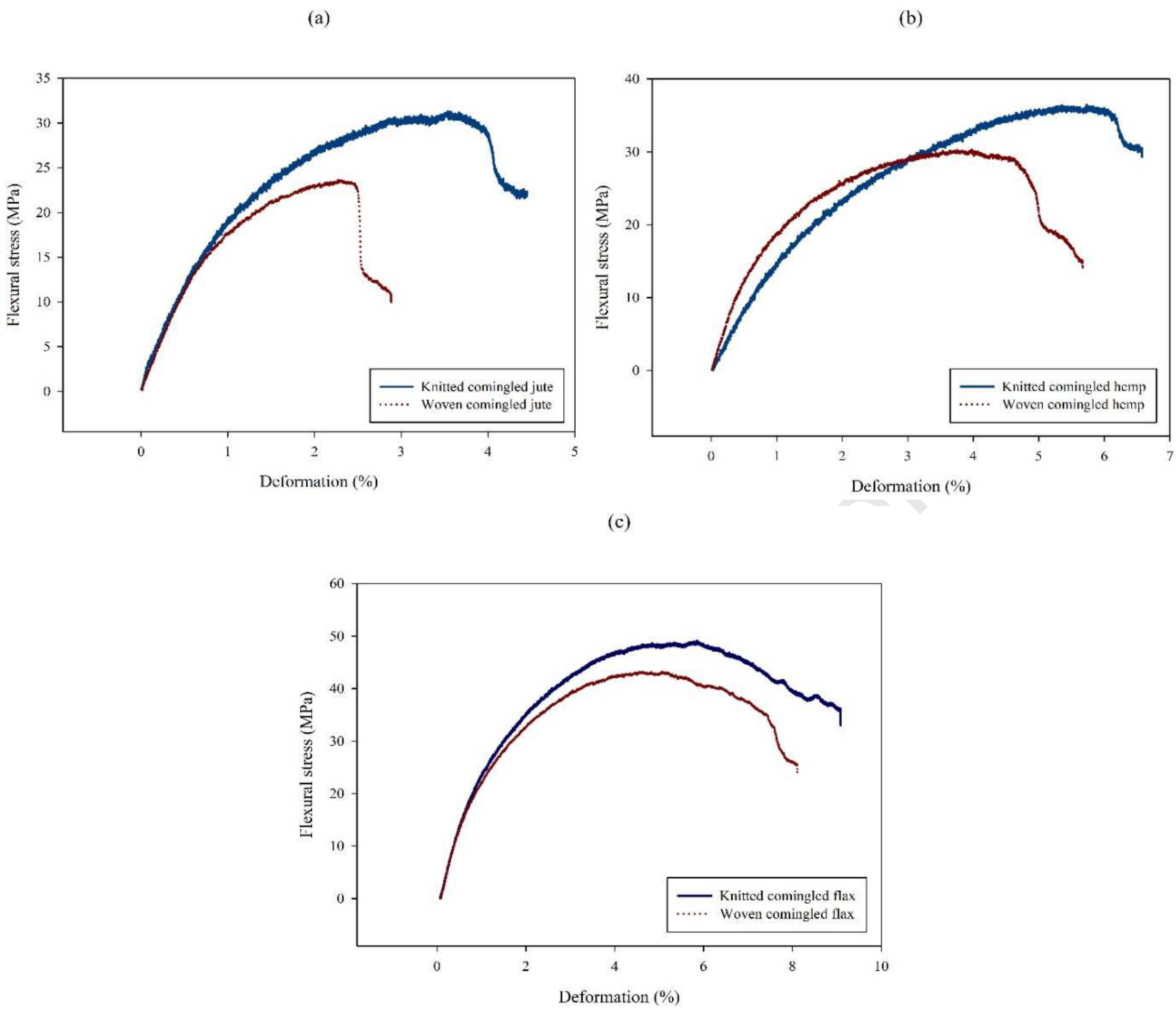


Fig. 8. Comparison of stress strain histories of 3-point bending of (a) jute, (b) hemp, and (c) flax woven and knitted comingled laminates

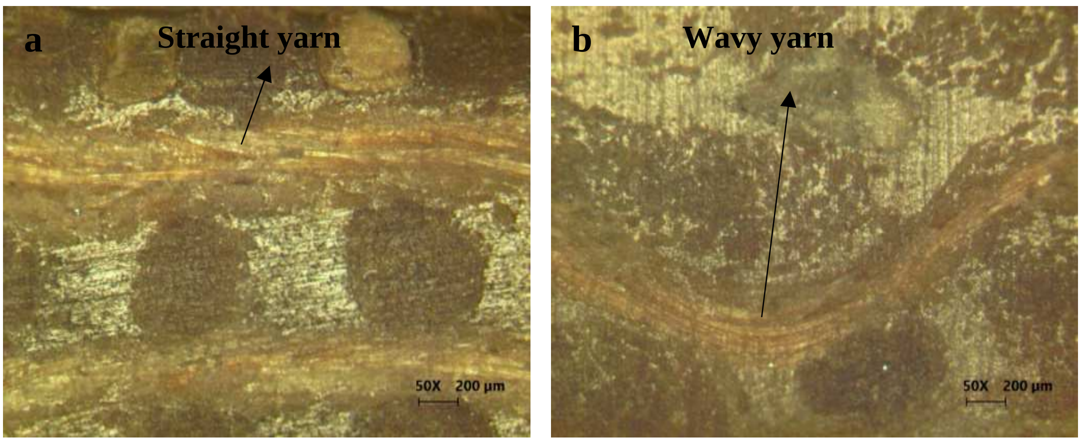


Fig. 9. Microscopic images of fibre waviness (a) knitted comingled composites (b) woven comingled composites

3.4. Impact strength

3.4.1. Drop-weight impact strength

Low-velocity impact is potentially dangerous, mainly due to the damage might be left undetected. These unseen damages can propagate under dynamic loading and even cause catastrophic failure to the structures. Composite laminates experiencing low-velocity impact damage often involve a combination of fibre fracture, fibre distortion, matrix cracking, fibre matrix de-bonding or delamination.

Rebounding, penetration and perforation are the three basic types of impact test cases. A closed force-deflection curve is indicative of the occurrence of rebounding phenomena [44]. It was observed that the composite samples did not experience any penetration at the impact energy level of 5 J as all the curves are closed contours (***Fig. 10***) and the incident energy was fully transferred to the specimen at the point of maximum displacement. The specimen then transferred the elastically stored impact energy back to the impactor after the maximum displacement. Afterwards, force curve returns to zero at the force-displacement curves. The maximum deflection values were summarized in ***Table 3***.

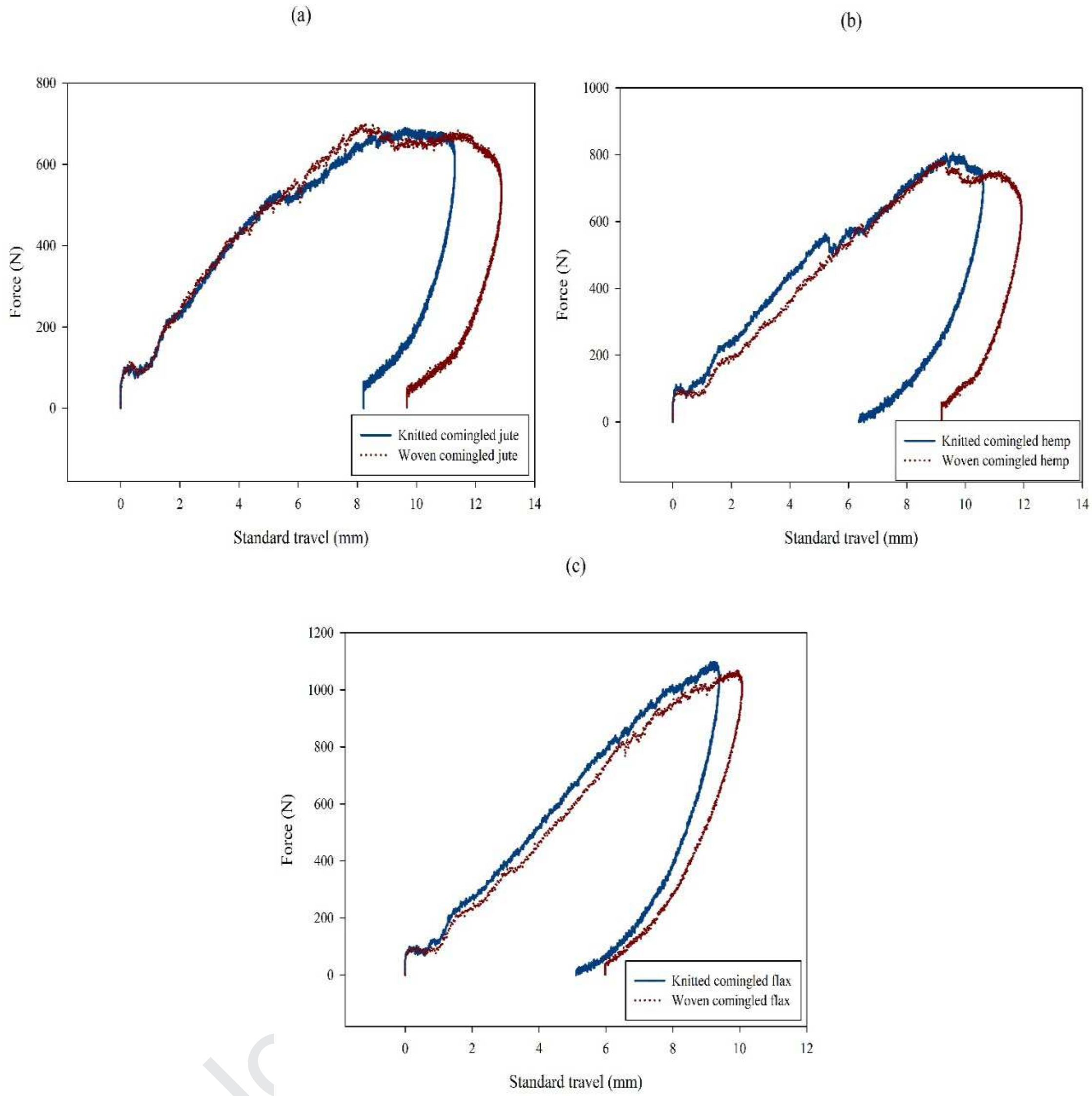


Fig. 10. Force displacement curves of drop weight impact specimens of woven and knitted comingled (a) jute, (b) hemp, (c) flax laminates

The deformation in the specimen was caused by the kinetic energy from the impactor and in accordance to the principle of conservation of energy, the energy absorbed by the specimen is equivalent to the energy taken up for damage creation. Therefore, a severely damaged specimen would have higher absorbed energy [45]. The absorbed energy values (***Table 3***) showed that knitted comingled composite specimens have lower energy absorption compared to the woven comingled composite specimens, which indicates that the knitted comingled composite specimen exhibit reduced damage formation compared to the woven commingled composite specimens at the same incident energy impact level. Therefore, it can be concluded that the knitted comingled composite specimens are more impact resistant than the woven composites. Furthermore, it can also be observed that the knitted comingled flax composite have the lowest energy absorption value, which proves that flax composites have better impact resistance of than the other remaining composite specimens.

Table 3

Comingling technique	Material	Maximum deflection (mm)	Absorbed energy (J)
Woven	Jute	12.87	4.69
	Hemp	11.92	4.51
	Flax	10.05	3.85
Knitted	Jute	11.29	4.46
	Hemp	10.60	4.22
	Flax	9.38	3.83

Maximum displacement of woven and knitted composites

3.4.2. Charpy impact test

The impact strength of the un-notched knitted comingled laminates was measured and compared with the woven comingled laminates. The results show that the Charpy impact strength had increased by 37 %, 54 % and 44 % for the jute, hemp and flax knitted comingled laminates (***Fig. 11***) compared to the woven comingled laminates respectively, which can be attributed to the reduced flow distance and increased wetting between the matrix and the reinforcing material. The improvement in fibre wetting helped to transfer the impact load within the composite. The composites made from flax comingled fabric were found to have higher impact strength than the hemp and jute composites due to the higher strength of flax yarn as compared to hemp and jute.

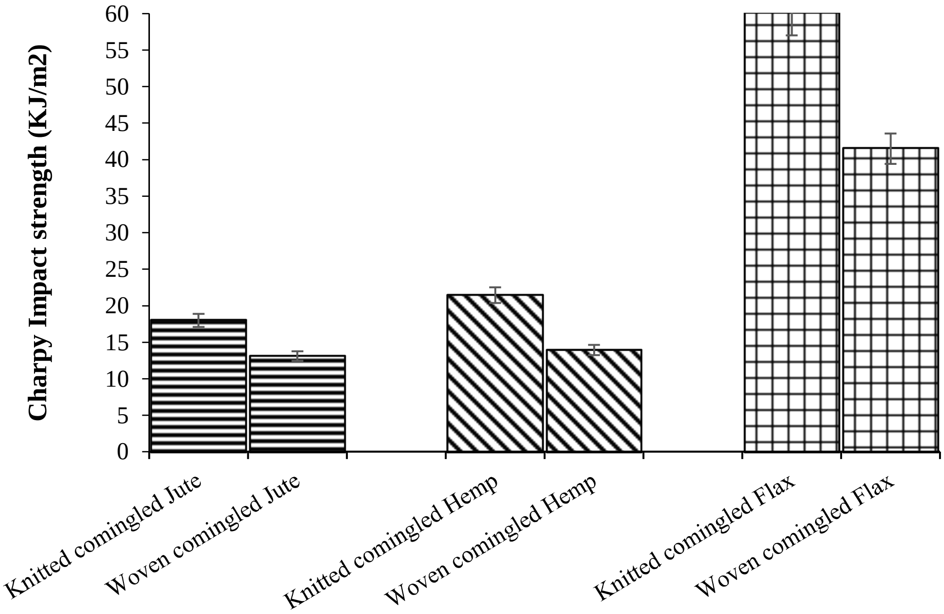


Fig. 11. Charpy impact strength of woven and knitted comingled laminates

4. Conclusion

Eco-friendly partially biodegradable and recyclable composites were developed to mitigate the environmental impact by using knitted and woven comingled reinforcements of natural yarns (jute, hemp, flax) and polypropylene yarns. The comingled fabrics as the reinforcing preforms were developed by keeping the count of yarns, area density, fabric density and the reinforcing yarns ratio constant to highlight the effect of the comingling technique on the mechanical properties of the resultant laminates. Knitted comingled composites exhibit better mechanical properties than the woven comingled composites. The tensile, flexural and impact properties of knitted comingled laminates were found to be higher compared to the woven comingled laminates. Furthermore, these properties for flax composites were superior to that of hemp and jute composites. These improvements can be attributed to the reduced flow distance between the matrix and reinforcement in the knitted comingled specimens, which enhanced the wetting/impregnation. Additionally, a maximum increase of 54 % was observed in the impact strength of the hemp knitted comingled composites.

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