Preparation and mechanical properties of novel bio-composite made of dynamically sheet formed discontinuous harakeke and hemp fibre mat reinforced PLA composites for structural applications

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Abstract

The objective of this study was to fabricate and assess the structure and mechanical performance of PLA reinforced by aligned discontinuous alkali treated natural fibre mats. The novel aspect of this work was in terms of composite processing, in particular, the use of fibre mats produced using a dynamic sheet former (DSF) wherein discontinuous treated fibres were aligned. Composites with different fibre weight contents were assessed using optical microscopy, scanning electron microscopy (SEM) and tensile testing. It was found that the fibres had a reasonable degree of alignment parallel to the DSF rotation direction together with good fibre dispersion. Tensile strengths of harakeke and hemp composites up to 101.6 MPa and to 87.3 MPa were achieved, approximately 90 and 60% higher respectively than PLA (53.9 MPa). It was also found that minimum and critical fibre volume fractions for harakeke and hemp fibre composites were lower than that found for injection moulded composites, due to better reinforcement efficiency obtained as a result of increased fibre alignment.

Keywords: Discontinuous reinforcement; Dynamic sheet former (DSF); Aligned fibre; Mechanical properties; Mechanical testing; Thermoplastic composites

1 Introduction

The potential of natural fibre as reinforcement in composite materials has been enhanced over recent decades due to improved mechanical properties as a result of improved technology used in preparing the reinforcing fibre and composites. Natural fibre composites are also recognised for their potential to reduce problems associated with petroleum based non-degradable composites. Plant fibres, in particular harakeke and hemp fibre are known to be amongst the strongest natural fibres. Harakeke is abundantly available in New Zealand commonly referred as ‘native flax’ used generally as clothing, mats, baskets and nets by the Maori people. Harakeke fibres are expected from leaves while hemp fibres are extracted from bast. It has been reported that tensile strengths of harakeke and hemp fibres were up to 800 and 1000 MPa respectively, at their untreated state. Some mechanical properties of harakeke and hemp fibres are shown in Table 1, along with E-glass for comparison (Aruan Efendy and Pickering, 2014; Dittenber and GangaRao, 2011; Zini and Scandola, 2011).

Table 1 Mechanical properties of natural and synthetic fibre.

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Density (g/cm³)</th>
<th>Length (mm)</th>
<th>Elongation (%)</th>
<th>Young’s modulus (GPa)</th>
<th>Tensile strength (MPa)</th>
<th>Specific tensile strength</th>
<th>Specific modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harakeke</td>
<td></td>
<td></td>
<td>4.2–5.6</td>
<td>14–33</td>
<td>440–990</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hemp</td>
<td>1.5</td>
<td>5–55</td>
<td>1.6</td>
<td>70</td>
<td>550–900</td>
<td>370–600</td>
<td>40</td>
</tr>
<tr>
<td>E-glass</td>
<td>2.5</td>
<td></td>
<td>2.5</td>
<td>70</td>
<td>2000–3000</td>
<td>800–1400</td>
<td>29</td>
</tr>
</tbody>
</table>

A number of car models, first in Europe and then in North America, have featured natural fibre composites in door panels, package trays, seat backs and trunk liners. In the construction industry, natural fibre reinforced composites are...
gaining popularity and used in non-structural applications such as for doors and window frame, wall insulation and floor lamination (Azeredo, 2009; Comradi et al., 2009; Kymäläinen and Sjöberg, 2008; Youssef et al., 2012). Generally, the performance of fibre reinforced composites depends largely upon the quality of individual fibres and matrix, strength of interfacial bonding between the fibre and matrix and the alignment of fibre; stress transfer at the fibre–matrix interface and fibre orientation are found to be major controlling factors for discontinuous fibre composites. It has been shown that a substantial improvement in fibre–matrix interfacial bonding can be achieved by utilising suitable fibre treatments and coupling agents. However, research on fibre orientation for discontinuous fibre composites is still limited. Discontinuous fibre composites made by either injection moulding or hot pressing randomly aligned fibre with polymeric powder or sheet are found to have low mechanical performance due to limited fibre length and poor fibre alignment. Composites reinforced with continuous fibre on the other hand have better mechanical properties due to higher reinforcement efficiency as a result of better stress transfer and fibre orientation, but their production is time consuming and limited to certain types of fibres. In this work, research efforts to improve fibre alignment for discontinuous fibre using a dynamic sheet former (DSF) was undertaken to improve the performance of discontinuous fibre composites. Although, it has been found that using DSF could improve fibre alignment of discontinuous fibre, there is no available data related to mechanical composite performance yet reported (Neagu et al., 2005). Harakeke and hemp fibres treated using alkali as developed in previous work were used (Aruan Efendy and Pickering, 2014).

2 Experimental

2.1 Materials

Harakeke fibre treated with 5wt% sodium hydroxide (NaOH) and 2wt% sodium sulphite (Na₂SO₃) and hemp fibre treated with 5wt% sodium hydroxide (NaOH) were used to produce fibre mats. The maximum length for both fibres was approximately 8 mm (cut using a granulator with 8 mm sieve size). NatureWorks® 3052D injection moulding grade PLA (polylactide) polymer with a density of 1250 kg/m³ from Nature Works LLC, USA was used as a thermoplastic matrix.

2.2 Methods

2.2.1 PLA sheet production

The manufacture of PLA and composite samples in this work involved converting PLA granules into PLA sheets. PLA granules were used as purchased and extruded into sheets of about 0.5–0.6 mm thick using an extruder equipped with a coat hanger die. The extruder consisted of 6 heating elements, for which the temperatures were set at 145 °C (barrel entrance), 165 °C, 180 °C, 180 °C, 170 °C (barrel exit) and 170 °C (die). The twin co-rotating screws were operated at 100 revolutions per minute (rpm). PLA sheets were cut to dimension of 150 mm × 90 mm to enable them to fit in a compression mould, with an approximate weight of 8–10 g each and kept in a sealed polyethylene bag.

2.2.2 Fibre mat production

Aligned fibre mats were produced using a Canpa ADSF dynamic sheet former (DSF) as shown in Fig. 1. In this study, fibre mats weighing 130–140 g/m² were produced using approximately 45 g treated discontinuous harakeke or hemp fibre. Firstly, fibre was dispersed in water (approximately 10 l of water was used to disperse 10 g of fibre) in a mixing drum fitted with a disintegrator. Mats were then produced by spraying a controlled quantity of this low concentration suspension inside a spinning drum containing a porous medium to retain the fibre using a controlled pump and drum spinning speed.

The fibre mats were finally rinsed and compacted by allowing the drum to spin for a further 2–3 min. They were then dried in an oven at 80 °C for 24 h before being cut to size (150 mm × 90 mm) to enable them to fit in a compression mould. Samples of dried harakeke and hemp fibre mats are shown in Fig. 2.
Randomly oriented fibre mats were produced by mixing fibres in water in a large container. The water containing the suspended fibres was allowed to slowly drain out of the container and the fibres were deposited on a strainer at the bottom of the container. Randomly oriented fibre mats were finally left to dry in the oven for at least 12 h before use.

### 2.2.3 PLA sample production

Fig. 3 shows the aluminium mould used to mould PLA and composite samples. The mould consisted of a base-plate onto which a central frame with internal dimensions of 150 mm × 90 mm was attached by screws to set the moulded plate width and length, as well as a T-shaped top section that could be inserted into the central frame, such that its flanges would rest on it to provide a gap of approximately 2.5 mm to determine the moulded material thickness. Small grooves were located at the bottom edges of the shorter sides of the central frame (as can be seen in Fig. 3c) to allow trapped air within the melt and the excess PLA to be squeezed out from the mould. PLA sheets were inserted in the mould in between Teflon sheets (see Fig. 3d) to give about 5–10% excess in weight relative to the calculated final weight of the moulded material. This was to ensure that the polymer melt flowed under some pressure and took the shape of the mould with minimum air bubbles.

PLA sheets were first preheated in the mould with no pressure at a temperature of 170 °C for 10 min. They were then compressed at 170 °C and 3 MPa pressure for 3 min. The mould was finally removed from the hot press and allowed to cool down to room temperature, with a low pressure applied by placing a steel block (10 kg) on the top of the mould, before the moulded PLA was removed and stored in a sealed polyethylene bag.

### 2.2.4 Fabrication of composite material

Fibre mats and PLA sheets were dried overnight at 105 °C and 60 °C respectively. They were weighed and arranged in a stack (in between Teflon sheets to prevent sticking to mould) with relative numbers of each based on the required fibre weight.
percentage following the arrangement as tabulated in Table 2 before inserting into the mould.

### Table 2 Arrangement of fibre mats and PLA sheets in mould.

<table>
<thead>
<tr>
<th>Approximate fibre wt%</th>
<th>Number of fibre mats</th>
<th>Number of PLA sheets</th>
<th>Fibre mat(s) and PLA sheet(s) arrangement</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>4</td>
<td>4PLA</td>
<td>4PLA</td>
</tr>
<tr>
<td>5</td>
<td>1</td>
<td>2PLA/1MAT/2PLA</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>2</td>
<td>1PLA/1MAT/2PLA/1MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>3</td>
<td>1PLA/1MAT/1PLA/1MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>4</td>
<td>1PLA/2MAT/1PLA/2MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>25</td>
<td>5</td>
<td>1PLA/3MAT/1PLA/2MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>6</td>
<td>1PLA/3MAT/1PLA/3MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>7</td>
<td>1PLA/4MAT/1PLA/3MAT/1PLA</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>8</td>
<td>1PLA/4MAT/1PLA/4MAT/1PLA</td>
<td></td>
</tr>
</tbody>
</table>

*Arrangement of PLA sheet and fibre mat from bottom to top of the stack; number = represent number of layer(s) used, PLA = PLA sheet and MAT = Fibre mat.

Stacks were heated and pressed in a hot press as for PLA samples (at 170 °C and pressed for 3 min at 3 MPa). Since fibre mat was easily distorted, consolidating PLA with fibre mats needed to be carefully conducted. It was ensured that PLA sheets were fully melted before slowly applying pressure; applying pressure onto insufficiently softened PLA sheets with viscosity that was too high distorted the fibre mats, which it was assumed would reduce composite strength. Air bubbles and excess PLA melt were squeezed out of the mould (through the grooves) as for PLA-only moulding. After hot pressing, the moulded composite materials were removed from the press and allowed to cool down (with a weight on top to apply pressure as for moulded PLA) to room temperature. Composite samples were then weighed to determine the final fibre weight percentage (from knowing the weight of fibre placed in mould) and stored in a sealed polyethylene bag. The fibre content of composite was increased from 5wt% until the fibre wetting become poor. Examples of moulded composites are shown in Fig. 4. It should be noted that, it was difficult to make smooth, rectangular uniform shape composites when the fibre content was higher than 30wt%.

### 2.2.5 Scanning electron microscopy (SEM)

SEM was used to observe surface morphology and fracture surfaces of tested specimens. The observations were conducted using a Hitachi S-4100 field emission scanning electron microscope operated at 5 kV. Samples were placed on aluminium

![Fabricated composites produced by compression moulding: (a) harakeke and (b) hemp composites with 20%wt fibre.](image-url)
stubs using double-sided adhesive tape and sputter coated with platinum and palladium to make them conductive.

### 2.2.6 Tensile testing of PLA and composites

In this work, all samples were cut into specimens of approximately 150 mm × 15 mm and their edges were ground to give a smooth and uniform section. Prior to tensile testing, all specimens were placed in a conditioning chamber at 23 °C ± 3 °C and 50% ± 5% relative humidity for at least 48 h. Tensile testing followed the procedures detailed in ASTM D 638-03: Standard Test Method for Tensile Properties of Plastics. It consisted of gripping a tensile test specimen in the jaws of an Instron-4204 tensile testing machine fitted with a 5 kN load cell; in order to prevent slippage and premature failure occurring near the grips, the specimen ends were cushioned with abrasive paper. 5 replicate samples were tested for each batch and average tensile strength (TS) and Young's modulus (YM) were obtained using the results from all specimens. An Instron 2630-112 extensometer was attached to the central part of the test specimen to measure the specimen extension. The specimens were tested at a constant rate of 2 mm/min. Details and abbreviations of tensile tested specimens used are tabulated in Table 3.

Stress versus strain graphs was obtained from which the ultimate tensile strength and Young's modulus could be determined; Young's modulus was obtained using the tangent to the initial linear portion of the stress-strain curve.

#### Table 3 Details and abbreviations of tensile tested composite samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Layer(s) of fibre mat</th>
<th>Approximate fibre wt%</th>
<th>PLA wt%</th>
<th>Load direction*</th>
</tr>
</thead>
<tbody>
<tr>
<td>PLA</td>
<td></td>
<td></td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>HR-5</td>
<td>HM-5</td>
<td>1</td>
<td>5</td>
<td>95</td>
</tr>
<tr>
<td>HR-10</td>
<td>HM-10</td>
<td>2</td>
<td>10</td>
<td>90</td>
</tr>
<tr>
<td>HR-15</td>
<td>HM-15</td>
<td>3</td>
<td>15</td>
<td>85</td>
</tr>
<tr>
<td>P-HR-15</td>
<td>P-HM-15</td>
<td>3</td>
<td>15</td>
<td>85</td>
</tr>
<tr>
<td>HR-20</td>
<td>HM-20</td>
<td>4</td>
<td>20</td>
<td>80</td>
</tr>
<tr>
<td>HR-25</td>
<td>HM-25</td>
<td>5</td>
<td>25</td>
<td>75</td>
</tr>
<tr>
<td>HR-30</td>
<td>HM-30</td>
<td>6</td>
<td>30</td>
<td>70</td>
</tr>
<tr>
<td>HR-35</td>
<td>HM-35</td>
<td>7</td>
<td>35</td>
<td>65</td>
</tr>
<tr>
<td>HR-40</td>
<td>HM-40</td>
<td>8</td>
<td>40</td>
<td>60</td>
</tr>
</tbody>
</table>

*Parallel and perpendicular relative to DSF rotation direction, HR = Harakeke and HM = Hemp.

### 2.2.7 Single fibre tensile testing

Single fibres were subjected to tensile testing according to the ASTM D3379-75: Standard Test Method for Tensile Strength and Young's Modulus for High-Modulus Single Filament Materials. Single fibres were mounted on 2 mm thick cardboard mounting-cards with a 2 mm gauge length as schematically shown in Fig. 5. PVA glue was applied to hold the fibres to the cardboard and define the gauge length. The diameter of the fibres was measured at five different points along the fibres length by means of an Olympus BX60F5 optical microscope. At least 20 replicate samples were tested for each batch and average tensile strength (TS) and Young's moduli (YM) were obtained using the results from all specimens.
3 Results and discussion
3.1 Assessment of fibre mat and composite morphology

Fig. 6 shows the optical images of randomly oriented fibre mats produced by allowing randomly dispersed fibre to settle from a suspension (a and b) and aligned fibre mats produced by the DSF (c and d).

It can be observed that in contrast to randomly oriented fibre mats, fibre mats produced by the DSF had fibres that were aligned to a reasonable degree parallel to the DSF rotation direction. It can also be observed that fibre mats produced using the DSF had improved fibre dispersion relative to randomly oriented fibre mat which could help to further improve the mechanical properties of composites. Fig. 7 shows composite surfaces, displaying fibre orientation within composites, while Fig. 8 shows cross-sections of composites parallel and perpendicular to DSF rotation direction. In Fig. 8, fibres sectioned longitudinally appear as dark lines whereas those sectioned transversely appear as light circles. Improved fibre alignment is supported as more transverse fibre cross-sections were observed within the sections cut perpendicular to DSF direction and more longitudinal fibre sections were observed within the sections cut parallel to the DSF rotation direction.
SEM micrographs of fracture surfaces of harakeke and hemp composites that were tested perpendicular (15 wt% fibre) and parallel (20 wt% fibre) to the DSF rotation direction are shown in Figs. 9 and 10. Fig. 9 further confirms that alignment of harakeke and hemp fibres has been obtained using DSF. It is evident that fibres were uniformly dispersed and well impregnated with PLA without obvious segregation of resin and fibre rich zones. Some holes in the matrix can be observed as well as protruding lengths of fibre (Figs. 9a and b) suggesting fibre pull-out has occurred, which could be related to poor interfacial strength or fibres having lengths shorter than the critical fibre length, leading to fibre debonding rather than fibre failure (Fu and Lauke, 1996). The gaps between fibres and matrix (shown in Fig. 10c and d) were smaller compared to untreated fibre composites observed elsewhere (Kabir et al., 2012b), suggesting a good degree of wetting known to be beneficial as a result of alkali treatment which would encourage good bonding. Furthermore, improved interfacial bonding is supported by longitudinal splitting of hemp fibres as seen in Fig. 9b, therefore suggesting the pull-out described above, being related to lengths of fibre shorter than the critical length rather than a weak interface.
3.2 Composite tensile testing—effect of fibre content and fibre orientation

Fig. 11 presents stress versus strain graphs for composites with 15wt% fibre tested parallel and perpendicular to the main fibre alignment direction along with that for neat PLA for comparison purposes. Both harakeke and hemp composites tested in both directions failed in a brittle manner at low strain without noticeable yielding. The stiffness for composites loaded parallel to the main fibre alignment direction (HR-15 and HM-15) was significantly increased as expected due to reinforcement and the failure strains remained comparable or increased (HR-15) relative to neat PLA. Increments in stiffness can also be observed in composites tested perpendicular to the main fibre alignment direction particularly for harakeke composite (very small though for hemp), but the failure strains for the composites loaded perpendicular to the main fibre alignment direction were significantly lower (Fischer, 2004; Graupner, 2009; Hu and Lim, 2007; Mathew and Joseph, 2007; Oksman et al., 2003).

Tensile strengths for harakeke and hemp composites versus fibre content are presented in Fig. 12. Clearly, for composites tested parallel to the main fibre alignment direction, the inclusion of fibres up to 30wt% for harakeke and 25wt% for hemp increased the tensile strength of the composites.
Maximum tensile strengths of harakeke and hemp composites were 101.6 MPa and 87.3 MPa respectively, which were approximately 90 and 60% higher than for PLA (53.9 MPa) with small standard deviations observed, as indicated by the error bars, suggesting good uniformity of fibre distribution throughout the composite. Higher improvement in tensile strength for harakeke composites could be due to a larger interfacial area as a result of smaller diameter fibres. It is believed that, higher improvement is also attributed to a better fibre separation for harakeke compared to hemp (see Fig. 6). It is evident that these factors outweighed the contribution of fibre strength; note that tensile strength of hemp fibre is higher than harakeke fibre (Aruan Efendy and Pickering, 2014).

Tensile strengths attained in this work with discontinuous fibre in PLA are better than those in other reported works with PLA and discontinuous fibre at similar fibre contents; this is assumed to be due to improved fibre alignment and conserved fibre length relative to other composites produced where extrusion and injection moulding have been used resulting in more random orientation and where lengths of fibres are generally reduced due to processing (Agari et al., 2007; Baghaei et al., 2013; Baghaei et al., 2014a; Baghaei et al., 2014b; Du et al., 2014; Graupner, 2009; Hu and Lim, 2007; Kabir et al., 2012a; Masirek et al., 2007; Plackett et al., 2003; Sawpan et al., 2011). Reduction of strength above 30wt% for harakeke and 25wt% for hemp could be due to insufficient polymer for adequate wetting and increased possibility of fibre–fibre interaction resulting in fibre agglomeration (Krishnaprasad et al., 2009), as well as an increase in the structural porosity component in the composite (Madsen and Lilholt, 2003). Furthermore, fibre was more easily displaced during processing at higher fibre contents due to the transverse pressure in polymer melt which could have contributed to a decrease in harakeke and hemp composite tensile strengths through reduced orientation and increased agglomeration.

Unsurprisingly, lower strength was obtained for composites tested perpendicular to the main fibre alignment direction. Although in the main fibre alignment direction, composite properties are strongly dependent of fibre properties and the interface, in the perpendicular direction, properties are more dependent on fibre–matrix interfacial bonding and the matrix (Baghaei et al., 2014b). Also, in this direction, the dimension (diameter) of the fibre is very small relative to the critical fibre length for shear stress to bring about tensile load in the fibre. Furthermore, it is likely that the strength of the fibre is lower due to orientation of microfibrils which has shown to give different properties in different fibre direction (Madsen and Lilholt, 2003).

Reduction in tensile strength for hemp composites with 5wt% fibre tested parallel to the fibre direction relative to PLA could be due to the fibre content being lower than that referred to as the critical fibre volume fraction for reinforcement. The concept of minimum and critical fibre volume fractions for unidirectionally-aligned continuous fibre composites is illustrated schematically in Figs. 13 ($V_{f,min}$) and 14 ($V_{f,min}$ and $V_{f,crit}$) for the two different possible cases such that the fibre has a higher or lower failure strain than the matrix (Hull and Clyne, 1996).
Typical stress–strain curves for harakeke, hemp and PLA in this work are shown in Fig. 15, showing that the material with the highest failure strain is harakeke followed by PLA followed by hemp with the lowest failure strain, so for harakeke composites the fibre failure strain is higher than the matrix and for hemp composites the fibre failure strain is lower.

In the case where failure strain of the fibre is higher than that for the matrix (Fig. 13 (ε_f > ε_m)), as the strain is the same in the fibre and the matrix during testing, the matrix fails first. At low fibre volume fractions, load is largely supported by the matrix and the fibre will not be able to support the load transferred from the matrix, so composite failure will occur when the matrix fails (matrix controlled failure); at higher fibre volume fractions, the fibre will be able to support the transferred load until the strain of the composite reaches the failure strain of fibre (fibre controlled failure). Ideally, (see Fig. 13) composite failure occurs when the fibre stress reaches σ_f, when the composite strength is given by σ_f V_f. The fibre volume fraction where the transition between matrix controlled failure and fibre controlled failure occurs is called the minimum fibre volume fraction (V_f,min). Above this point an increased gradient in composite strength versus fibre fraction occurs as the reinforcement becomes more efficient, until it reaches maximum fibre volume fraction (V_f,max). Beyond V_f,max the composite strength deteriorates due to significant increase in porosity, poor wetting and inefficient stress transfer as a result of limited fibre–fibre spacing (Shah et al., 2012). For the case where the fibre has a lower failure strain than the matrix (Fig. 14 (ε_f < ε_m)), which is the situation for hemp fibre, at low fibre contents, as the strain on the composite surpasses the fibre failure strain, the fibre fails first leaving the matrix unreinforced with the fibres effectively acting as holes (matrix controlled failure). Consequently, tensile strength of a composite reduces with increased fibre content and is a minimum at V_f,min. As the fibre content increases beyond V_f,min, when the fibres fail, the composite fails (fibre controlled failure), however, with more fibre, more load is carried and so the strength increases and strength greater than that of the matrix can be seen to be obtained at a fibre volume fraction higher than that defined as the critical fibre volume fraction.
Beyond $V_{\text{f,min}}$, the maximum stress on the matrix ($\sigma'_m$) is dependent on the failure strain of the fibre.

Fig. 16 shows the experimental data for tensile strength as a function of fibre content as previously presented in Fig. 12, with lines fitted to enable minimum, critical and maximum fibre volume fractions to be obtained. From Fig. 16a, it can be seen that $V_{\text{f,min}}$ and $V_{\text{f,crit}}$ for harakeke are about 10 and 32% respectively. The $V_{\text{f,min}}$, $V_{\text{f,crit}}$ and $V_{\text{f,max}}$ for hemp are about 4.5, 6 and 22% respectively, for hemp (Fig. 16b). Furthermore, $\sigma_f$ (defines the fibre stress at composite failure strain) for harakeke fibre is found to be approximately 358 MPa and $\sigma'_m$ (defines the matrix stress at fibre failure strain) is found to be approximately 40 MPa. Clearly, for the case of hemp composites, strength greater than the strength of PLA can only be expected if the fibre volume fraction is higher than 6%. Notably, the $V_{\text{f,min}}$ and $V_{\text{f,crit}}$ in this work are lower compared to those reported for other short fibre composites. For instance, the $V_{\text{f,min}}$ and $V_{\text{f,crit}}$ for short banana fibre reinforced vinyl-ester were found to be around 15 and 25% respectively (Ghosh et al., 2011), while for hemp fibre reinforced unsaturated polyester composites were around 20 and 40% respectively (Sawpan et al., 2013). This indicates that composites reinforced with these aligned natural fibre mats have better reinforcement efficiency as a result of better fibre alignment and interfacial properties. However, the values obtained from this work are still considerably higher compared to those obtained for aligned carbon-polyester composites which were found to be around 2.3 and 2.4% (Shah et al., 2012).

Young's moduli of harakeke and hemp composites tested parallel to the fibre direction are shown in Fig. 17. It can be seen that Young's modulus improved linearly with increasing fibre content up to 30 and 25wt% for harakeke and hemp fibre composites respectively. This is expected due to the fact that fibre possesses higher Young's modulus than PLA (Graupner, 2009).
Improvement in Young’s moduli for harakeke and hemp were approximately 120% (8.02 GPa) and nearly 165% (9.67 GPa) at 30 and 40wt% respectively, compared to that for PLA (3.6 GPa). It may be seen that at the same fibre contents, Young’s moduli for hemp composites are higher than that harakeke composites, reflecting the higher Young’s modulus for hemp fibre (26.33 GPa) compared to harakeke (21.2 GPa). However, for composites tested perpendicular to the fibre direction (for 15wt% fibre content), Young’s modulus for harakeke composite is found higher compared to hemp composite; suggesting that Young’s modulus for harakeke in this direction could be higher or possibly higher interfacial bonding between the harakeke fibre and the matrix as a result of smaller fibre diameter and/or better fibre separation. In contrast to tensile strength, Young’s modulus for composites tested perpendicular to fibre direction were found to have increased. This trend has been also found elsewhere (Baghaei et al., 2014b; Joseph, 1999), which suggests that Young’s modulus is less dependent on fibre orientation and fibre/matrix interface but more on fibre content, a higher fibre content providing increased constraint within the matrix.

Failure strains for harakeke and hemp composites are shown in Fig. 18. As can be seen, failure strains for harakeke composites tested in the main fibre alignment direction were not greatly affected by the inclusion of fibres unlike for hemp composites for which failure strain reduced as fibre content increased. A greater reduction in failure strain for hemp fibre composites compared to harakeke fibre composites is not surprising given the lower failure strain of hemp fibre (1.58%) relative to that of harakeke fibre (3.95%). Lower failure strain for hemp composites could also be due to more fibre agglomerates and a higher void content particularly at higher fibre fractions resulting in increased stress concentration as reported elsewhere (Pickering, 2008).
For composites reinforced perpendicular to the test direction (both harakeke and hemp), significant reduction in failure strains is noticed when compared to the failure strains of composites tested in the fibre direction, suggesting that the stress concentration as a result of fibre inclusion in this direction encourages matrix cracking to occur at strain level lower than the failure strain of PLA only.

4 Conclusion

The alignment of fibres within composites reinforced by fibre mats produced using a DSF and the tensile strength of PLA composites reinforced using these mats with different fibre contents tested parallel and perpendicular to the fibre direction was investigated. It was found that better fibre orientation and dispersion was obtained using a DSF than by allowing the fibre to randomly settle from a suspension. A reasonable interface between fibre and matrix appears to have also been obtained as a result of alkali treatment. Reinforcing PLA using fibre mats produced by a DSF was found to be an effective technique to improve mechanical properties. Maximum tensile strengths for harakeke and hemp fibre composites were found to be 101.6 MPa and 87.3 MPa respectively (approximately 90 and 60% respectively higher than that of PLA). Young's modulus of harakeke and hemp fibre composites were also improved by nearly 120% and 160% respectively compared to PLA. It was also found that the minimum and critical fibre volume fractions were lower compared to composites made using injection moulding as a result higher reinforcement efficiency through fibre alignment obtained using a DSF.

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**Highlights**

- Discontinuous plant fibres were successfully used as reinforcements in PLA matrix.
- Dynamic sheet former (DSF) successfully aligned the fibres to a reasonable degree.
- Improved fibre orientation greatly improves tensile properties of composites.
- Tensile strength for harakeke composite at 20wt% fibre was 101 MPa.

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**Queries and Answers**

**Query:** The author names have been tagged as given names and surnames (surnames are highlighted in teal color). Please confirm if they have been identified correctly.

**Answer:** Checked and confirmed

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