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Developing and characterizing new materials based on waste plastic and agro-fibre

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Abstract This study optimizes flexural properties of composites made from waste materials, corn stalk, seed flax and *Agave americana* fibres along with waste linear low-density polyethylene (LLDPE) and high-density polyethylene (HDPE) matrices. The surface morphology of fibres and composites was characterized with light and scanning electron microscopes. Thermal behaviour of the corn stalk outer rings and pith parenchyma was shown using thermogravimetric analysis. There was no significant difference between the LLDPE composites made with corn stalk outer rings versus whole stalks, possibly due to an insignificant role played by parenchymatous part of the pith. Water retted seed flax fibres in LLDPE matrix optimized the composite flexural strengths at 6–8 days of retting, above which the properties declined. Field retted seed flax varieties in LLDPE matrix showed no significant differences in their flexural strengths. With additional fibre loading, flexural strength of *A. americana* HDPE composites first dropped before improving after extrusion, and only improved when using the layer method.

Introduction

The driving force behind natural fibre composites (NFCs) is that natural fibres are low in cost, sustainable, environmentally friendly and abundantly available in most world regions [1]. One way to take advantage of these many benefits is to focus on underutilized agricultural residues that normally end up as waste. The selection of fibre type is then based on the specific availability in each region. Some underutilized fibres include corn (maize) stalk, seed flax, and *Agave americana* fibres. Further, the use of local waste polymers instead of virgin ones can provide further economic and environmental benefits to NFCs. Although a good work has been done on the area of NFCs, the current literature is limited with respect to their production using waste starting materials, and hence this topic forms the basis of this study.

Studies of corn stalk in composites are beginning to surface. Ganjyal et al. [2] obtained corn stalk fibres by immersing the stalks in sodium hydroxide. The fibres were then used to reinforce starch acetate matrices and were used as packaging material. The shear strength of the composites improved with increasing fibre content, which could be attributed to good compatibility between starch acetate and corn stalk fibres. Panthapulakkal and Sain [3] used milled corn stalks to reinforce high-density polyethylene matrix (HDPE). HDPE reinforced with corn stalks did not improve much in mechanical properties and this was attributed to a weak interface between the filler and matrix. However, mechanical properties of the corn stalk HDPE composites were comparable to those of HDPE reinforced with commonly used wood flour.

Corn stalk has a heterogeneous structure [4]. The outer ring is a concentration of stronger fibres bound by pectin and lignin. Despite a few fibres forming part of vascular bundles, the pith has mainly a cellular parenchymatous tissue which would presumably act chiefly as a filler rather than reinforcement in composites. It would be assumed that separating the outer ring from the pith and reinforcing with this ring alone could make stronger and stiffer composites than if the whole stalk was used. However, if there is no significant difference in properties between using the entire

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stalk compared with just the outer ring, then the composite production process becomes more efficient and cost-effective as the separation step could be eliminated. The authors are not aware of any study that has examined the influence of using the whole stalk versus the outer ring on the mechanical properties of corn stalk thermoplastic composites; this became the subject of one of our studies.

Compared to corn fibres, flax fibres have recently received a fair amount of attention from researchers; however, the major focus has been on fibre flax (flax grown solely for the fibre) rather than seed flax (flax grown for the seed). The successful use of seed flax could provide further environmental and cost benefits as they are already extremely abundant at the end of seed harvest and have no current applications. A necessary process in flax fibre production is retting, which makes use of microorganisms to free fibres from stems. In water retting, a bacterium, which is naturally available in surrounding air, water and soil, degrades the pectin that binds fibres to the woody stem [5]. Fungus is mainly responsible for field retting where straw is left on the ground for some extended periods. Composites from retted fibres are predicted to have superior properties as retting removes gummy pectinic substances (impurities) that cause stress concentrations in composites [6]. Furthermore, the freed microfibres are able to orient themselves in the direction of the applied force, which allows them to bear greater loads.

Retting can be performed for various periods of time where the degree of retting has an influence on the properties of natural fibres [7]. Over-retted fibres begin to lose strength due to excessive degradation of load bearing cellulose, whereas under-retted fibres are not able to achieve the purpose of retting. Currently, the direct effect of degree of retting in terms of time duration on the mechanical properties of the composites is not well known. Such knowledge would give a clearer picture of what constitutes optimum retting times and could help decrease mechanical variability between composites. Another aspect of flax is that seed flax fibres come in many different varieties and it is necessary to know which varieties give the strongest composite properties. These aspects have been focused on in our study.

Although an advantage of NFCs is the local availability of natural fibres in most regions of the world, many fibres that have a strong potential for reinforcement have not been sufficiently studied. *A. americana* fibres are a good example [8–11]. In some parts of the world, *A. americana* plants are potentially the biggest source of strong but underutilized fibres [8, 12]. For instance, in Lesotho, Southern Africa, these fibres are a waste product of the processing of skin lotion products by local cooperatives, although they were traditionally used for making ropes and hats [12]. To our knowledge there are no studies that show the direct influence of these fibres on the mechanical properties of thermoplastic composites. Some of the unique properties of these fibres may help dictate the choice of composite processing materials and methods for many parts of the world [8, 10].

Finally, it has been shown that NFCs are environmentally superior to glass fibre composites particularly in automotive applications mostly due to weight reduction. However, they may not necessarily be superior in other applications like housing [1, 13]. The use of waste polymers instead of virgin ones can enhance the environmental benefits of NFCs in housing and other applications. Polymers that are widely used such as polyethylene are more attractive.

In this study we demonstrate how flexural properties of composites from waste materials can be optimized with respect to cost, environment and regional availability. This is done through a careful selection of relevant fibres, matrices and processing parameters. The products are targeted at both a developed (Canada) and a developing (Lesotho) country. The objective is to develop materials products that could be used in building and furniture applications and would be able to withstand a substantial load.

Experimental

Materials

Corn stalks, seed flax straw and *A. americana* leaves were the sources of fibres used in this study. Chemical contents of fibres from these sources are shown in Table 1. Corn stalks were obtained from local farmers. In order to examine the influence of the use of the outer ring versus the whole stalk on the composite flexural properties, the outer ring was easily peeled from the pith using knives. Both the outer rings and the whole stalks were cut to pieces small enough to feed into the milling machine. These samples

Table 1 The chemical content of corn stalk, seed flax and *Agave americana* fibres

Fibre	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Ash (%)	Reference
Corn stalk	38–40	7–21	28	3.6–7.0	[14]
Seed flax	43–47	21–23	24–26	5	[15]
Agave americana	66–72	10–14	12	–	[16]

Table 2 The characteristics of the fibres used in this study

Fibre	Diameter[a]	Length/mesh size (cm)		Tensile strength (MPa)	Fibre/Flour extraction
		Layer	Extrusion		
Corn	N/A	N/A	0.2[b]	N/A	Milling
Flax	30–105	N/A	1–3	345–1500 [17]	Retting, decortication, scutching
Agave americana	80–210	4–15	1–3	120–260 [8]	Hot water treatment

[a] Measurements of 40 samples per fibre types were made using Clemex Vision image analysis software, assuming cylindrical shape of fibres

[b] Mesh size for corn stalk flour

were then milled separately with Thomas Wiley laboratory mill using a mesh size of 2 mm.

Seed flax fibres came from Biolin Research Inc. Six different types of seed flax straw were used: Vimy seed flax with samples water retted in non-distilled tap water for a period of 0, 2, 4, 6, 8, 10 and 12 days at room temperature. Flanders, Omega, Nuggets, Evelyn and Hermes seed flax were field retted for 121 days (took much time because weather conditions were less conducive). The straws were then passed through a decorticator to extract the fibre, which was cleaned by scutching.

Agave americana fibres were sourced from Lesotho. Fibre extraction involved heating the leaves in boiling water for 2 h until they were soft [8]. The fibres were then removed by gently squashing the leaves with rocks and washing away the soft tissue with running water. They were then left in the sun and wind by hanging them along the fence until they were dry. Certain characteristics of the fibres used in this study and their respective extraction process are summarized in Table 2. Before use, all the fibres or flour were dried in an oven at 60 °C for 24 h to reduce moisture.

The HDPE plastic bags produced by Hymopack Ltd. (similar to those used as commercial packing bags in Lesotho) were sourced from the waste stream. The linear low-density polyethylene (LLDPE) films, which were produced by AEP Industries Inc. and used as waste bale wrap, were obtained from local farmers. All the plastic films were washed using a commercial dish washing liquid to remove the dirt. They were then dried in an oven for 24 h at 60 °C. The rationale behind material selection is summarized in Table 3.

Methods

Corn stalk/seed flax LLDPE composites

Corn stalk flour and LLDPE films were weighed to achieve desired weight fractions (Table 3). Then the flour was evenly laid between several films of LLDPE (up to three films on both sides of the flour) and hot pressed for a few seconds between two films of Teflon sheets (by CS Hyde Company) at 150 °C and a pressure just great enough to pre-impregnate the fibres into the matrix (Fig. 1). Nearly the same ratios of plastic and flour were used per composite film made to maintain uniform distribution of flour per weight fraction. This resulted in thin composite films which were chopped using scissors into nearly square pieces of about 1 × 1 cm to make their feeding into the extruder easier. The pieces were then taken into the Wayne single screw laboratory extruder for further compounding. There were five heating zones on the extruder which were set at 140, 150, 160, 170 and 180 °C, with the highest temperature (180 °C) at the last heating zone towards the end of the barrel. The screw speed was set at 15 rpm. The compounded composites came through a circular die that resulted into long cylindrical composites with a diameter of around 6 mm. These composites were then cut into pellets of around 1–2 cm in length. The pellets were put into a 12.7 cm × 12.7 cm × 3.2 mm stainless steel mould and again hot pressed between Teflon sheets at 3 MPa and 150 °C for 3 min into approximately 3.2-mm-thick sheets ready for cutting and testing (Fig. 2).

To make the seed flax LLDPE composites, the long seed flax fibres were first chopped into 1–3 cm long fibres using scissors. Then the composite production followed the same process described for the corn stalk LLDPE composites above.

The *A. americana* HDPE composites

The production of *A. americana* HDPE composites followed two methods. Firstly, the composites were made using the process detailed in the previous section. The second method (layer method) involved using the *A. americana* fibres of the length 4–15 cm (Table 2). These longer fibres were laid between thin films as illustrated in Fig. 1 and then hot pressed as in the previous section to make thin composite films (Fig. 2). Nearly the same ratios of plastic and fibre were used per composite film made to

Table 3 The materials, processing, experimental parameters and the rationale behind their choice

Target country/ Rationale	Fibre	Plastic	Method	Parameters	Weight fraction (%)
Canada	Corn stalk	LLDPE	Extrusion	Influence of corn stalk outer ring vs. whole stalk	0, 10, 20, 30, 40, 50
Rationale	Corn stalk: abundant agricultural waste in Canada (i.e. in Ontario and Québec [18, 19]). LLDPE: abundant recyclable polymer used in Canada as bale wrap		Affordable in a developed country	Using whole stalk could save processing costs	Little knowledge on the influence of weight fraction for corn stalk fractions
Canada	Seed flax	LLDPE	Extrusion	Influence of retting duration/fibre variety	20
Rationale	Seed flax: abundant agricultural waste in Canada (i.e Saskatchewan and Manitoba) [20]		Affordable in a developed country	Optimizing properties while avoiding costs of over-retting. Varietiy may have effect on properties	Influence of flax fibre loading in thermoplastic is well studied; keeping it constant helps in measuring other vital variables
Lesotho	*Agave americana*	HDPE	Extrusion/layer	Influence of processing method	0, 5, 10, 20, 30
Rationale	*Agave americana*: abundant but less used source of strong plant fibres in Lesotho [8, 12]		The layer method could be cheaper for developing countries	A cheaper processing method that could also optimize properties would be ideal	Little knowledge on fibre loading influence could not go beyond 30% due to too high viscosities

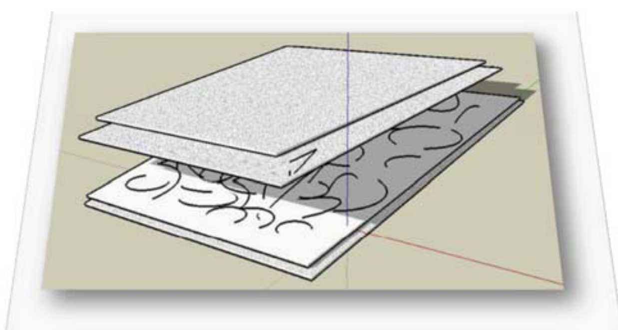


Fig. 1 The layering of fibre and plastic films to make thin composite sheets for further compounding or hot pressing

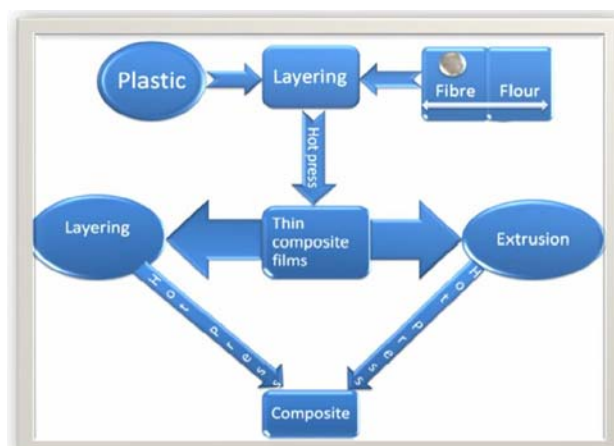


Fig. 2 The stages and processes of making the composites

maintain uniform distribution of fibres per weight fraction. After hot pressing, the roughly 13 × 13 cm composite films made were again layered together in a 12.7 cm × 12.7 cm × 3.2 mm stainless steel mould and hot pressed under the conditions mentioned in the previous section (Fig. 2). The products were approximately 3.2-mm-thick sheets ready for cutting and testing. The weight percent of these composites is shown in Table 3.

Flexural tests

The flexural tests were performed through the three-point bend tests using an Instron 3369 machine following ASTM D790-97. The three-point bend test method was specifically developed for testing flexural properties of unreinforced and reinforced plastics and it has been commonly used for testing composites similar to the ones in this study [3, 21]. Whereas the three-point bend tests may underestimate flexural modulus, it is often a preferred method over four-point bend test since it requires less material for each test and does not need the determination of centre point deflections [21]. The method was considered to be acceptable for this study as it is mainly comparative. The specimens for testing were cut by machining, making five specimens per variable. The dimensions of each test specimen were kept at approximately 127 × 12.7 × 3.2 mm. The support span was 51.2 mm, using the span to depth ratio of 16:1.

The rate of crosshead motion R was 1.37 mm/min, calculated using Eq. 1,

$$R = \frac{ZL^2}{6d} \quad (1)$$

where L is support span (mm), d is the depth of the beam (mm), Z ($Z = 0.01$) is the rate of straining of the outer fibre (mm/mm/min). The stress in the outer fibres at mid span, σ (MPa) or flexural strength was taken when the strain reached 0.05. It was calculated using Eq. 2:

$$\sigma = \frac{3PL}{2bd^2} \quad (2)$$

where P is load (N), L is support span (mm), b is width of the beam (mm) and d is the depth of the beam (mm). The flexural modulus E (MPa) was calculated using Eq. 3:

$$E = \frac{L^3 m}{4bd^3} \quad (3)$$

where L is the support span (mm), b is the width of the beam (mm), d is the depth of the beam (mm) and m is the slope of the tangent to the initial straight line portion of the load-deflection curve (N/mm).

Microscopy

In order to determine the influence of processing on the morphology of the fibres and the composites, two kinds of microscopes were used to characterize these materials. Olympus stereo light microscope connected to a camera was used to capture the surface morphology of the composites and corn stalks. For finer morphological details, samples were first gold plated and then examined under the scanning electron microscope. The process used to extract single fibres of *A. americana* for SEM analysis is found in [10]. It involved heating the *A. americana* fibres in 1 N solution of sodium hydroxide at 130 °C for 300 min in an autoclave (Autoclave engineers Inc.). The process breaks down the lignin binding the single fibres without destroying the fibres.

Thermogravimetric analysis

Small samples of the parenchyma (with the vascular bundle fibres removed) and the outer ring were put in platinum pans and analyzed with a thermogravimetric analysis machine (TA Instruments Inc.) in a nitrogen atmosphere. The heating rate was 10 °C/min from 25 to 700 °C. The results were analyzed using TA Instruments Universal Analyzer 2000 software.

Results and discussion

Corn stalk LLDPE composites

Few studies show a potential of corn stalks in reinforcing thermoplastics. Therefore it was necessary to vary fibre loading and to investigate both the strength and moduli of the composites in addition to variables of concern (Table 3). Corn stalk is made of a woody fibrous outer ring consisting of epidermis and mainly peripheral vascular bundles [4, 22]. It has an inner tissue consisting of vascular bundles and parenchyma. The tensile strengths of corn stalk fibres are not well documented in literature. However, the more fibrous outer ring should add more stiffness and strength to the matrix than the more cellular inner tissue. Therefore separating this ring from the center of the stalk should result in better composite flexural properties than if whole stalks were used. The results of this comparison are shown in Figs. 3 and 4.

High standard deviations are a characteristic for the properties of both natural fibres and materials based on them. The whole corn stalk composites reached the maximum flexural strength at 30% fibre loading (47%

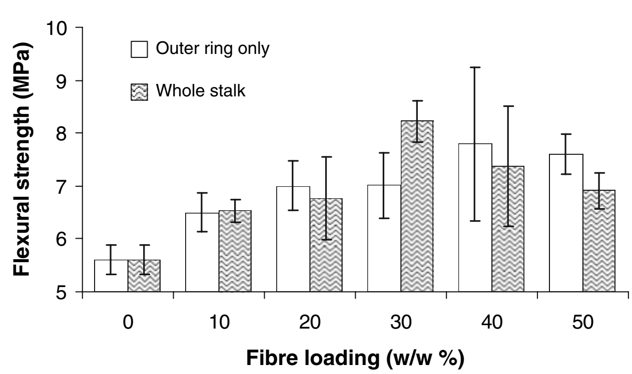


Fig. 3 The flexural strengths of LLDPE reinforced with the corn stalk outer rings and the whole stalks

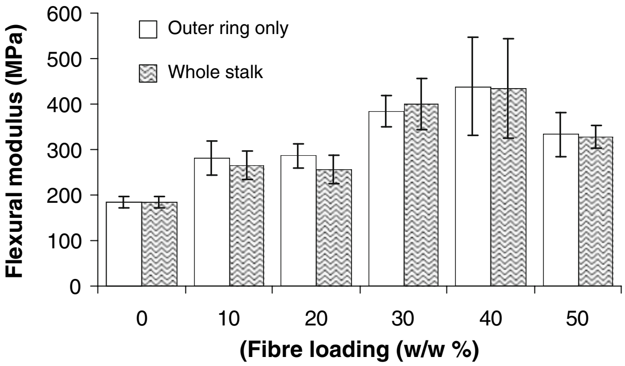


Fig. 4 The flexural moduli of LLDPE reinforced with the corn stalk outer rings and the whole stalks

improvement compared to pure LLDPE) and then declined a little from 40% fibre loading. The outer ring corn stalk composites reached maximum flexural strength at 40% fibre loading (39% improvement compared to pure LLDPE). However, the two reinforcements did not differ significantly at each fibre loading. Corn flour could be expected to improve the properties moderately. In addition to their weaker strengths, milled corn particles come in many shapes. This results in a reduced area to volume ratio and therefore less contact area between the filler and the matrix. This is in contrast with flax or *A. americana* fibres which, in addition to their greater strengths, have a higher aspect ratio (length to diameter ratio) and therefore larger contact area between fibre and matrix per volume of fibre added.

There are possible reasons for the small differences between the two composites in Figs. 3 and 4. The most apparent would be the weight distribution in the corn stalk. The outer ring consists of densely glued fibres which make up the majority of the weight of the stalk (Fig. 5). For instance, a piece 11 cm long with a diameter of roughly 3.4 cm cut from the stalk had only 30% of its weight as the pith. That weight included both the parenchyma and vascular bundle fibres. Consequently, the percentage weight of the parenchyma compared to the whole stalk is relatively small, or according to Hooper [22], negligible. Therefore, the minor influences of the parenchyma on composite properties, along with the presence of stronger fibres in the pith, result in little change when using the entire stock compared to just the outer ring. In both composites, the modulus was almost doubled at 40% fibre loading although the two composites remained almost the same at all fibre loadings (Fig. 4). The decline in flexural properties at higher fibre loading could be due to poor dispersion as the amount of corn stalk in the plastic increased. Hydrophilic corn stalk particles interact more with themselves as their content increases, resulting in poor interface [23, 24].

Thermal degradation effects on corn composites

Moreover, thermal degradation analysis showed that whatever little contribution the light parenchyma might have on the strength or modulus, it could be maintained beyond processing temperatures. The thermal analyses

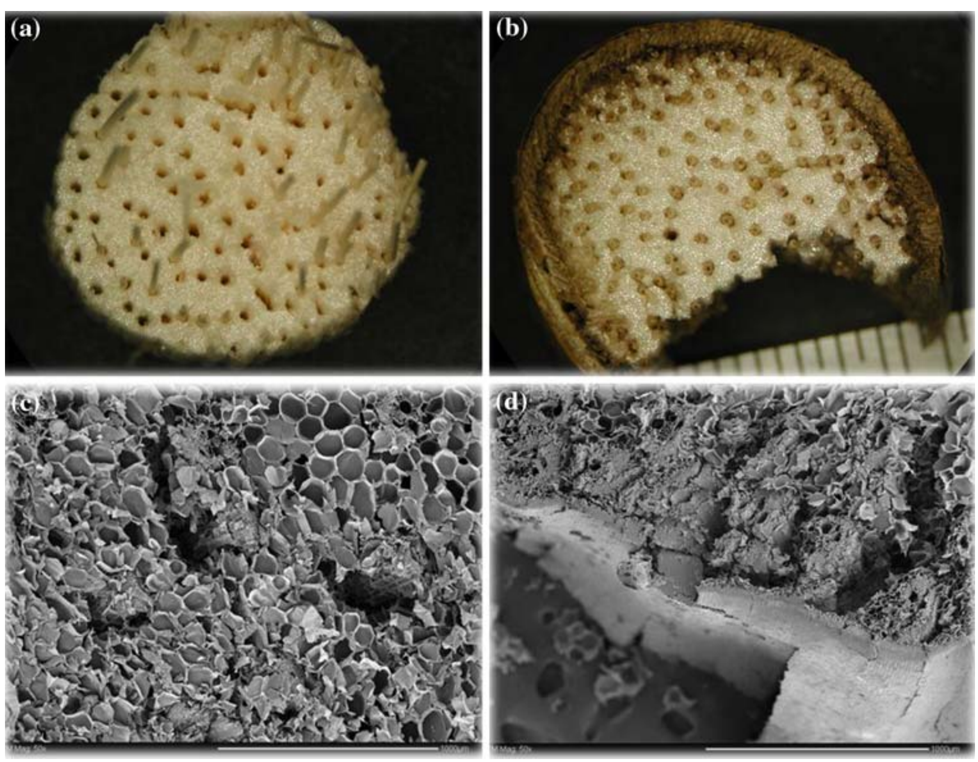
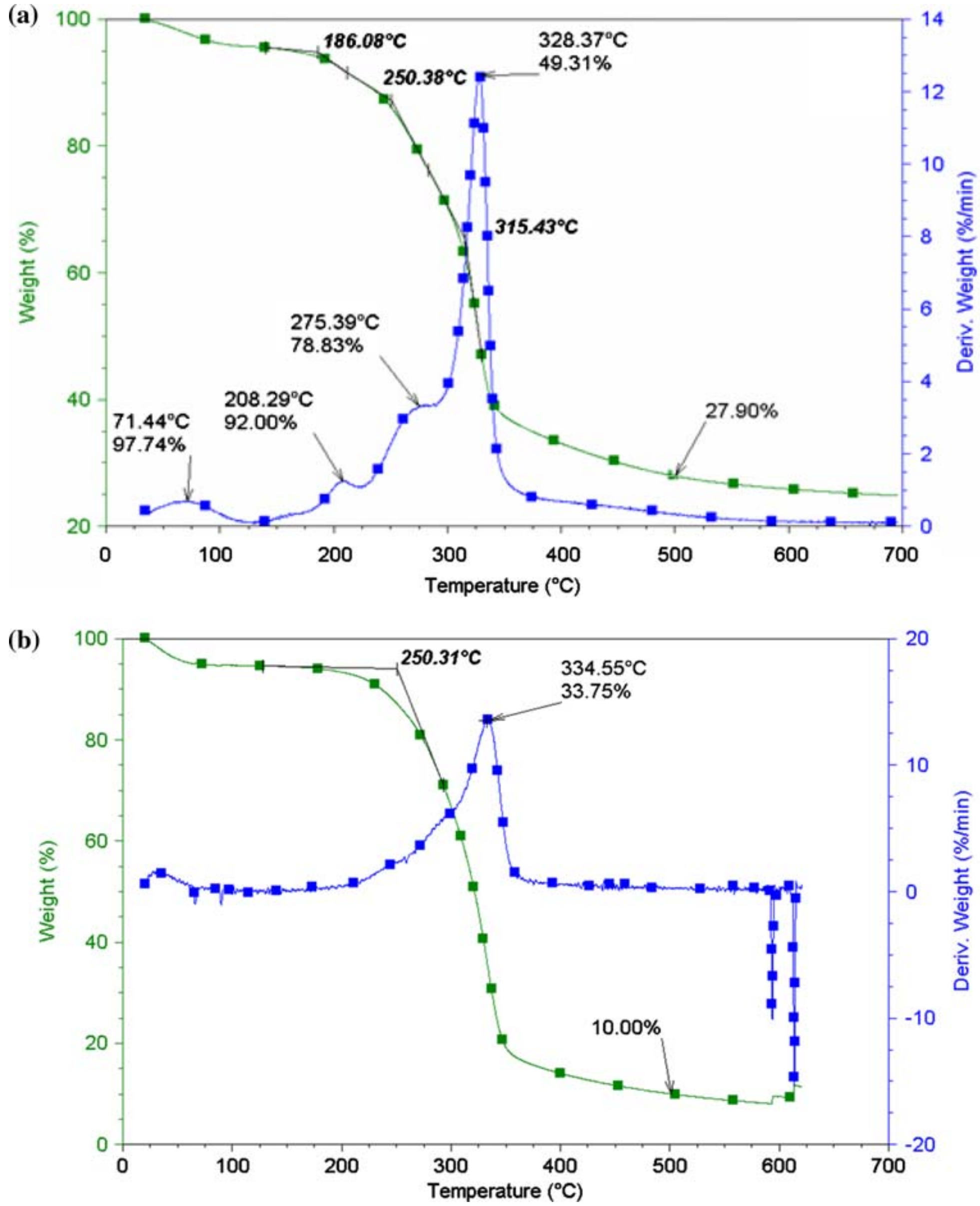


Fig. 5 (**a**) Light microscope pictures of corn stalk pith showing spongy parenchymatous tissue with some vascular bundles in the form of fibres distributed within the tissue. (**b**) A picture of corn stalk pith surrounded by woody outer ring which consists of peripheral vascular bundles and epidermis. A bar on the scale represents 1 mm. (**c**) An SEM picture (50×) of a cross section through corn stalk pith showing walls of dead parenchymatous cells. (**d**) An SEM picture (50×) of a cross section through corn stalk showing the woody outer ring and a region of peripheral vascular bundles

Fig. 6 The thermal behaviour of (a) the outer ring and (b) parenchyma



showed that the parenchyma began to degrade at 250 °C, well above the highest composite processing temperature of 180 °C (Fig. 6a, b). In contrast, the outer ring had some components that began to degrade at around the processing temperatures (186 °C).

Whereas the parenchyma had only one significant peak at 335 °C signaling the degradation of cellulose, the outer ring had several peaks at 208, 275 and 328 °C, likely signaling the degradation of pectin, hemicelluloses and lignin and cellulose, respectively. The curve in Fig. 6a may imply that the parenchyma is chiefly cellulose due to little evidence of other peaks. The first peak below 100 °C for both corn stalk components is due to dehydration. At 500 °C, the outer ring and the pith had 27 and 10% of the weight remaining, respectively. Higher residues in the outer ring might be due to higher lignin and mineral content in this component [3].

Influence of fibre retting duration and fibre type on seed flax LLDPE composites

The primary purpose of retting is to make fibre separation (decortication) easier by reducing energy consumption and other costs otherwise used during the process. Besides this primary function, retting adds an additional benefit to composites by cleaning the fibre. Figure 7a, b shows the SEM images of unretted and 12 day water retted Vimy seed flax technical fibres (a fibre bundle of small single fibres), respectively. The unretted flax fibre has some

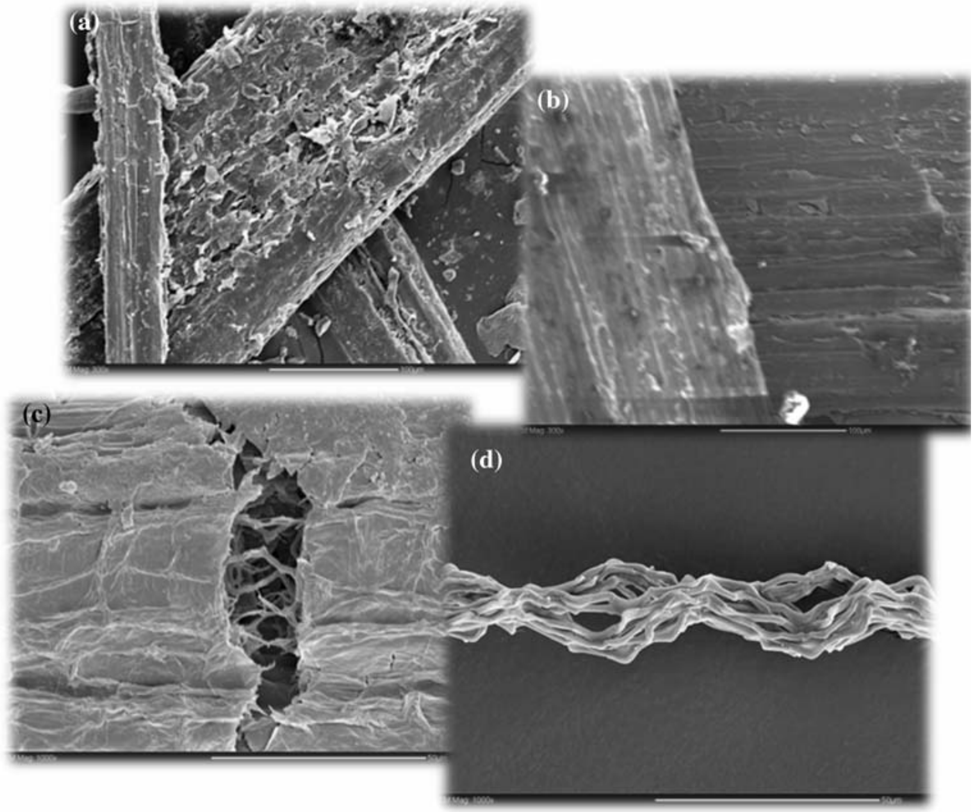


Fig. 7 (**a**) An SEM picture (300×) of unretted seed flax technical fibre showing gummy pectinic substances on the surface. (**b**) An SEM picture (300×) of a relatively smooth 12 day water retted seed flax technical fibre. (**c**) An SEM picture (1,000×) of the surface of a technical fibre of *A. americana* which has been extracted by immersing *A. americana* leaves in boiling water. Cracks on the surface reveal presence of minute ultimate fibres of the diameter ranging from 1.5 to 3 µm. (**d**) An SEM picture (1,000×) of a group of single fibres of *A. americana* showing a zigzag shape of these fibres

pectinic materials on its surface (Fig. 7a). The retted flax shows a relatively smooth surface (Fig. 7b).

The seed flax LLDPE composites were manufactured using extrusion (Fig. 2). The pure LLDPE matrix had a flexural strength of 5.6 MPa. Unretted flax fibre at 20% fibre loading improved the flexural strength of the matrix by 98% to 11.1 MPa (Fig. 8). The degree of retting had some influence on the flexural properties of the composites. The flexural strength increased from 11.1 MPa for unretted to 13.4 MPa after 8 days (21% improvement). But the flexural strengths declined after 8 days of retting until they reached a low of 9.9 MPa after 12 days of retting. This was

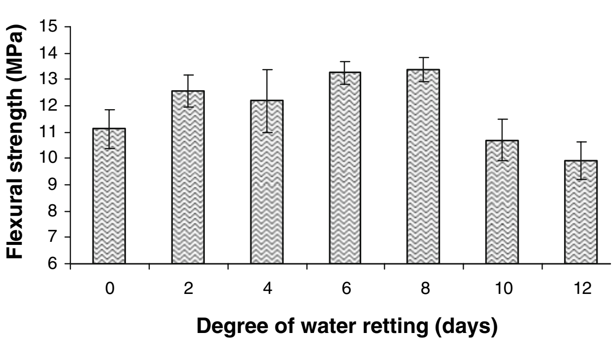


Fig. 8 The flexural strengths of extruded Vimy watter retted seed flax fibre/LLDPE composite at 20% fibre weight

a 26% percent decline from the peak at 8 days of retting. At this time (12 day duration), the strengths were also 11% below those of the unretted seed flax composites.

Retting has the capacity to remove impurities that cause stress concentrations which propagate cracks during application of force on the composites [6]. Also, as the pectin, lignin and hemicelluloses are degraded by the bacteria, the freed, mainly cellulose fibres can bear load better. They are better oriented towards the direction of load. Dijon [25] noticed that unretted flax fibres had poorer tensile properties than the retted ones. He argued that this was because unretted flax fibres were subjected to more mechanical force, therefore more damage, during decortication, and Van de Weyenberg et al. [6] gave similar reasoning.

However, it also known that natural fibres can be under-retted or over-retted [7]. It is important to closely define these terms especially on the direct influence of these possibilities on the composites. This knowledge can inform fibre processors concerning how far the retting process could go. Based on the conditions of this study (section "Experimental"), the optimum number of days for water retting Vimy seed flax fibre was between 6 and 8 days. However, the results may vary with seed flax variety and different fibre processing conditions.

In Fig. 9, flexural strengths of the composites reinforced with different field retted seed flax varieties are compared. The higher standard deviations are noticeable on this graph

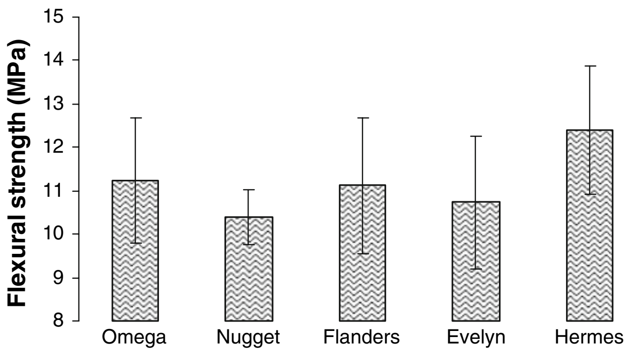


Fig. 9 The flexural strengths of extruded seed flax/LLDPE composites using different varieties of field retted seed flax fibres at 20% fibre weight

compared to results from water retted seed flax composites (Fig. 8). Field retting exposes flax straw to a variety of inconsistent localized biological and weather conditions, which results in inconsistent fibre properties [5]. These inconsistencies can be reflected on the composite properties. There are some variations between the flexural properties of the composites from these fibres. The noticeable difference of 1.9 MPa is between Hermes and nugget seed flax fibres. Omega, Flanders and Evelyn fibres are not much different. The generally small differences leave farmers with a large choice of fibres to grow.

The *A. americana* HDPE composites

The SEM images of *A. americana* show that its typical technical fibre is made up of hundreds of zigzag-shaped microfibres (Fig. 7c, d) [8]. The surveyed literature does not show any studies of composites reinforced with *A. americana*. Therefore it was necessary to vary fibre loading and investigate both the strength and moduli of the composites in addition to the targeted variables (Table 3). The two methods of production, layering and extrusion, lead to different results for *A. americana* HDPE composites (Figs. 10 and 11). Pure HDPE had flexural strength of 15.8 MPa using the extrusion. Upon reinforcement the flexural strength dropped down to 11.7 MPa (26% drop) at 10% fibre loading, but began to improve at 20% fibre loading and even more so at 30% fibre loading. Assuming a reasonable interface, the properties should increase with increasing fibre content.

The initial loss in strength in Fig. 10 is likely due to the use of pellets (section "Experimental"). Figure 12a, b shows what might be the cause of poor strengths in the extruded *A. americana* HDPE composites. The distinct unmerging pellets can be seen on the pictures. These distinct pellets in a composite are in contrast to the homogenous surfaces of the seed flax LLDPE and the corn

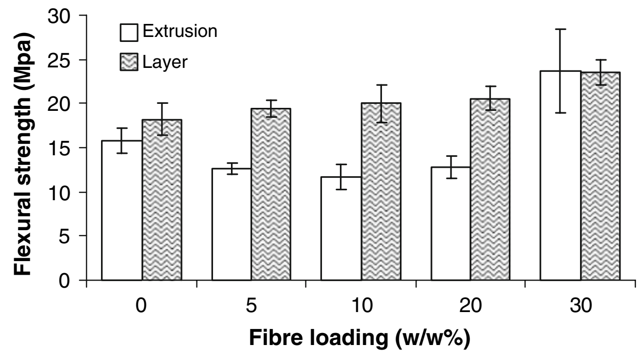


Fig. 10 The flexural strengths of *A. americana* HDPE composites of different processing methods

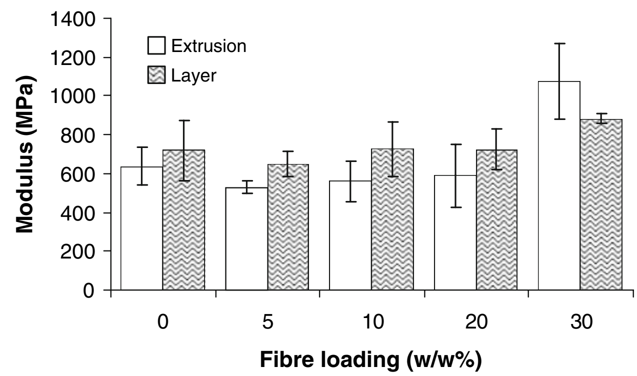


Fig. 11 The flexural moduli of *A. americana* HDPE composites of different processing methods

stalk LLDPE composites (Fig. 13a, b). HDPE has a higher viscosity and therefore lower melt flow index of around 0.3 g/10 min compared to LLDPE with the melt flow index of 0.7–1.3 g/10 min [26, 27]. Also the influence on viscosity of adding fibre to thermoplastics can be likened to the influence of reducing temperature [28]. For instance, due to high viscosity of these composites, the extruder could not handle fibre loadings above 30% as a result of the high pressures needed to convey them. High viscosity due to both the polymer and addition of fibre likely reduced integration of flows where the pellets met during hot pressing. The unmerging pellets could result in lines of weaknesses between them which reduced composite strength. The outcome seems to resemble the phenomenon of weld lines that can occur from injection molding where polymer flows from opposing directions meet. Further investigations could explain better why the properties improved at higher fibre loadings.

If the use of pellets is a problem as Fig. 12a, b indicate, the process could be optimized. By changing the extruder die, the extruded materials could come out not as long cylindrical pellets but as flat rectangular sheets that could be placed against each other as in layer method and hot pressed (section "Experimental").

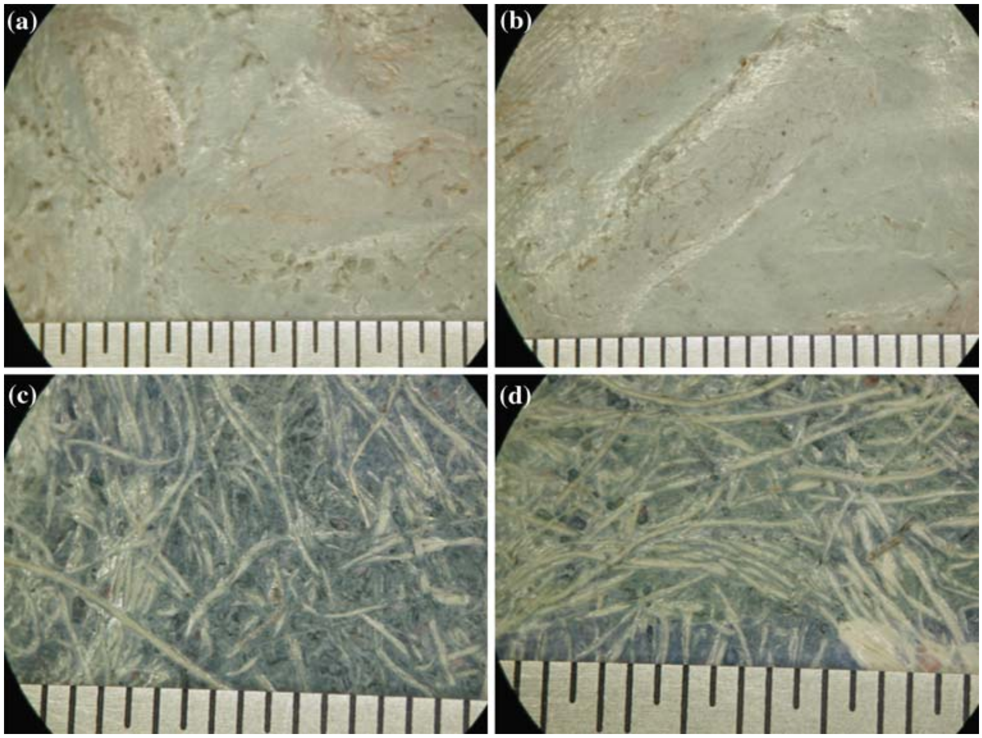


Fig. 12 (**a**, **b**) Light microscope pictures of the surface of *A. americana* HDPE composites hot pressed from extruded pellets. Some several millimetre thick pellets are evident with no signs of fully merging. (**c**, **d**) The surfaces of the *A. americana* HDPE composites made using the layer method. They show randomly distributed fibres on the horizontal plane of the composite. Each bar in the scale represents 1 mm

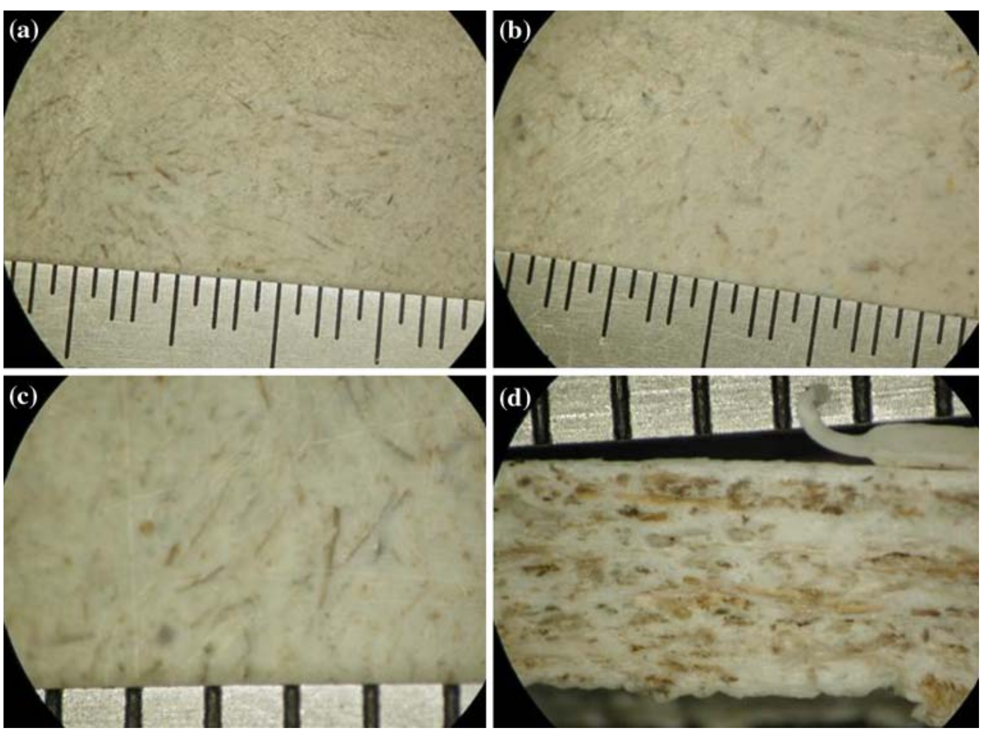


Fig. 13 (**a**, **b**) Light microscope surfaces of seed flax LLDPE composites and corn stalk respectively, made using the extrusion (20% w/w for both). (**c**) Cross section through the beam showing better fibre distribution in extruded seed flax LLDPE composites. The fibres are more randomly oriented in all possible directions and more encapsulated in a matrix. (**d**) Layered seed flax LLDPE composites. Fibres are not fully encapsulated; the fibres are oriented mainly on the horizontal plane of the composite. A bar on the scale represents 1 mm

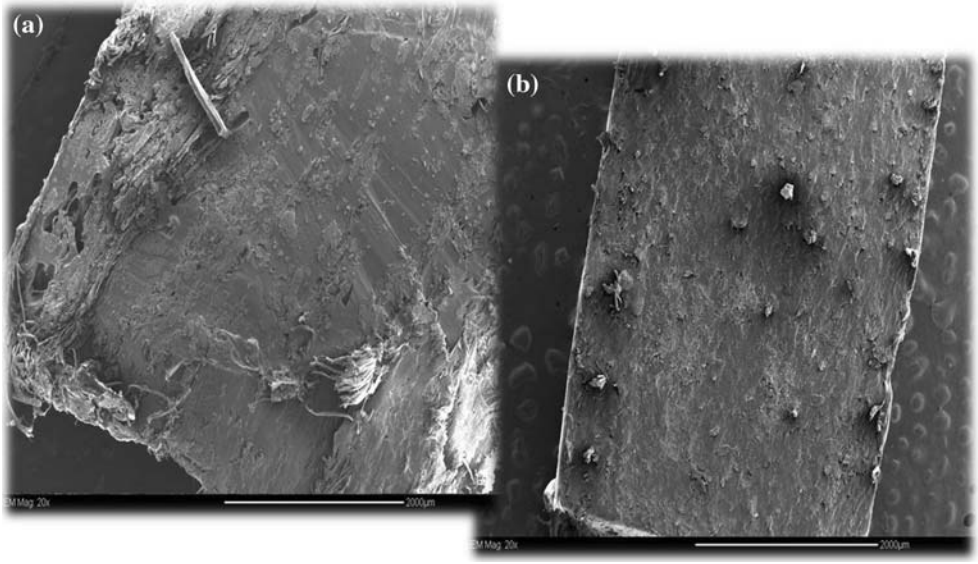


Fig. 14 (**a**) An SEM picture of the cross section in a seed flax fibre LLDPE composites using the layer method 20×. (**b**) An SEM picture of the cross section in a seed flax fibre LLDPE composites using the extrusion 20× (20% w/w for both)

The use of the layer method did not show the same problem for *A. americana* HDPE composites (Figs. 10 and 12c, d). With this method, the flexural strengths improved with increasing fibre content, reaching 29% improvement at 30% fibre loading compared to pure HDPE. This shows that there could have been weak lines present in the extruded samples that are clearly not there with the layer method. The modulus did not vary much with fibre loading except for the highest loading (Fig. 11). Also, Fig. 11 shows that except at 30%, the layer method has a little higher modulus values than the extrusion.

Although the layer method does not result in good mixing (Figs. 13d and 14a, b), the fibres in this method are randomly oriented mainly along the horizontal plane as opposed to all possible directions (Fig. 12c, d and compare Fig. 13c, d). Being better oriented could help them bear load better.

Conclusion

In this study we demonstrate how flexural properties of composites from waste materials can be optimized with regards to cost, the environment and the regional availability of materials. This is done through a careful selection of relevant fibres, matrices and processing parameters.

The presence of pith in corn stalk LLDPE composites does not make a significant difference in the flexural properties compared to when only the outer ring is used. Even though the pith takes a large volume of corn stalk, it consists of very light parenchymatous tissue and strong vascular bundle fibres which together make its influence on the composite properties relatively insignificant. Therefore, bypassing the process of separating the pith from the outer ring will decrease costs and energy use without compromising the composite properties.

Under the experimental conditions of this study, 6–8 days of water retting for Vimy seed flax fibre produced optimum composite properties. The fibre is possibly overretted beyond this period as shown by the decline in properties. This result can inform processors concerning how far the retting process could go, thereby saving the costs arising from over retting while optimizing the composite properties. Further, the relative similarity between flexural strengths of composites produced from different seed flax varieties shows that seed flax could be an abundant source of natural fibre for the NFCs industry.

Lastly, the *A. americana* HDPE composites lost and recovered their flexural strengths in response to increased fibre loading when they were extruded, pelletized and hot pressed. High viscosity of the composites could result in poor merging of pellets during hot pressing. In contrast, the same composites showed improvements at increasing volume fractions when the layer method was used.

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