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'Green' composites Part 2: Characterization of flax yarn and glutaraldehyde/poly(vinyl alcohol) modified soy protein concentrate composites

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In the present research, soy protein concentrate (SPC) was modified using glutaraldehyde (GA) and polyvinyl alcohol (PVA). The modified resin allowed to process soy protein polymer without any plasticizer. The modified resin also showed increased tensile properties, improved thermal stability and reduced moisture resistance as compared to SPC resin. Besides the tensile and thermal properties, modified SPC resin was also characterized for its dynamic mechanical properties.

Unidirectional composites were fabricated using modified SPC and flax yarn. Composite specimens, approximately 1 mm thick, were prepared in both longitudinal and transverse directions. The composite specimens were characterized for their tensile and flexural properties. The fracture surface of the composite was also analyzed in both longitudinal and transverse directions. These composite specimens exhibited a fracture stress of 126 MPa and 2.24 GPa, respectively, in the longitudinal directions. The composite properties were also predicted using the rule of mixture in longitudinal direction. It was observed that the experimental values are lower than the predicted values for a variety of reasons.

1. Introduction

Fully biodegradable, environment friendly, 'green' composites manufactured using plant based fibers and biodegradable resins offer an attractive alternative to conventional petroleum based plastics. Considerable research efforts have been made to develop soy protein based 'green' composites [1–9]. A brief review of various soy protein modifications is presented elsewhere [6, 10]. Part 1 of this paper discussed the soy protein concentrate (SPC) modification using glutaraldehyde (GA) and the thermal, mechanical and physical property changes as a result of the modification [10]. 'Green' composites using modified SPC and flax fabrics and their mechanical and flexural properties have also been discussed [10]. These 'green' composites showed good tensile and flexural properties and may be used for packaging or indoor paneling [10].

SPC polymer, processed without any modification, has low tensile properties, poor moisture resistance and is brittle [7]. For this part 2 of the research SPC was modified using GA and polyvinyl alcohol (PVA) to increase its tensile and thermal properties, moisture resistance and improve its processability as a resin to fabricate flax yarn reinforced composites. In addition, PVA being a good bonding agent, the fiber/resin bonding was expected to improve as well. SPC has been shown to form a miscible blend with GA and to react with free amine groups in the protein leading to a cross-linked structure with increased tensile properties [10]. Studies on SPC/PVA blend showed that blending PVA with SPC resulted in increased moisture resistance for the modified resin [11]. Hence modifying SPC with both PVA and GA was expected to improve the tensile properties as well as decrease the moisture content of the modified resin and make it more suitable as a resin for composite fabrication.

In this research spun flax yarns were used for fabricating composites. Flax fibers come in lengths of about one meter. As a result, these fibers cannot be used in continuous form. Use of twisted yarns in the composites, in such cases, allows the fabrication of continuous length unidirectional composites using short staple flax fibers. Mohanty *et al.* [12] fabricated biodegradable composites using twisted jute yarn and Biopol™. They reported that jute yarn reinforcement resulted in more than 150% increase in the tensile and impact strengths of the resulting biocomposites as compared to the pure resin. In the case of filament yarns, only low twist is required to keep them from spreading [13]. However, in the case of short staple (length) fibers,

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higher twist level is necessary to prevent fiber slippage and to develop sufficient strength. In addition, twisting localizes the micro damages within the yarn leading to higher fracture strength [14]. Spun yarns also have some fibers protruding out, commonly referred to as yarn hairiness. Although not desirable in many cases, the hairiness can lead to better mechanical yarn/resin interlocking in composites. Another advantage of using twisted yarns in composites is the increased surface roughness of yarns compared to fibers, which again increases the interfacial strength due to mechanical interlocking, improving the transverse properties.

The twist affects the yarn strength significantly [15]. Besides yarn strength, the amount of twist also affects the resin penetration while fabricating yarn reinforced composites. With increased twist yarns become more compact making it difficult for the resin to penetrate, leading to lower bonding between the fibers and the resin. Very low interfacial bonding can lead to delamination and lower the composite tensile properties.

2. Experimental procedures

2.1. Materials

ARCON® S, SPC powder was provided by Archer Daniels Midland Company, IL. Analytical grade glycerin and sodium hydroxide were obtained from Fisher Scientific, PA. GA, 25 wt% solution in water, and PVA, 98% hydrolyzed, with an average molecular weight (M_w) of 13,000–23,000, were obtained from Aldrich Chemical Company, Milwaukee, WI. All chemicals were used as received, without any further treatment. Flax yarn, in bleached form, was provided by Sachdeva Fabrics Pvt. Ltd., New Delhi, India.

2.2. SPC curing and modification

In the research presented here, SPC was modified with both PVA and GA. SPC powder was processed to make it suitable as a resin for 'green' composite fabrication using the similar process as described earlier in part 1 of this paper [10]. Desired amount of PVA was dissolved in distilled and deionized water with continuous stirring using a magnetic stirrer, in a water bath maintained at 80°C. The SPC powder was mixed with PVA solution in 1:15 ratio and glycerin was added as a plasticizer. The solution was homogenized using a magnetic stirrer for 15 min and the pH was adjusted to 11 ± 0.1, by adding 1 N NaOH solution. The SPC/PVA/glycerin solution was stirred for additional 15 min and then transferred to a water bath maintained at 70°C. The solution was pre-cured at 70°C for 27 min and then 40% (w/w of SPC) GA was added. This amount of GA (25 wt% solution) was determined to be optimum [10]. The SPC/PVA/GA blend solution was further stirred for 3 min while in water bath.

After pre-curing, the solution was cast on to Teflon® coated glass plates and dried in Fischer Isotemp oven at 35°C for 15–20 h. Finally, the dried modified SPC sheets were hot pressed (cured) in Carver Hydraulic hot press at 120°C for 25 min under a pressure of 7 MPa. The cured polymer sheets were conditioned at standard atmosphere (65% r.h. and 21°C) for 3 days before performing various characterization tests.

2.3. Effect of glycerin content

Glycerin has been shown to be an effective plasticizer for soy protein [3, 7, 10, 16, 17]. To study the effect of plasticizer content on the tensile properties of GA and PVA modified SPC (MSPC-GP) resin, four different glycerin contents, 0, 5, 10 and 15%, were used. Tensile properties of MSPC-GP polymer were characterized in accordance with ASTM D 882-97. Conditioned polymer sheets were cut into rectangular specimens of 110 × 20 mm dimensions. Three thickness measurements were made along the length of each specimen, and the average of those values was used for calculating the fracture stress and Young's modulus. The tests were performed on an Instron tensile tester, model 1122, at a strain rate of 1 min^{-1} and a gauge length of 50 mm.

2.4. Thermogravimetric analysis of SPC and MSPC-GP

Thermogravimetric properties of conditioned SPC and MSPC-GP polymer sheets were measured using TA Instruments, Thermogravimetric Analyzer (TGA), model 2050. The specimens were scanned in nitrogen atmosphere from 25 to 350°C at a ramp rate of 10°C/min.

2.5. Dynamic mechanical analysis of SPC and MSPC-GP

Dynamic mechanical properties of SPC and MSPC-GP polymer sheets were measured using TA Instruments Dynamic Mechanical Analyzer (DMA), model 2980, at Cornell Center for Materials Research (CCMR) facilities, using tension film clamps. Conditioned polymer sheets were cut into rectangular specimens of 11 × 6 mm dimensions. The tests were performed at 5 μm amplitude and 1 Hz frequency. The specimens were scanned from 0 to 240°C at a ramp rate of 5°C/min for both SPC and MSPC-GP polymers.

2.6. Characterization of flax yarn

Flax yarn was characterized for its diameter and tensile properties such as Young's modulus and fracture stress and strain. Yarn specimens were conditioned at standard ASTM atmosphere for 5 days, prior to characterizing their properties.

Flax yarn diameter was determined using Bausch and Lomb optical light microscope. Due to the large variation in natural fibers and their yarns, twenty specimens, each 75 mm in length, were used to measure their diameters. Three measurements were made along the length of each specimen and the average of these values was used for calculating the mean yarn diameter.

Tensile properties such as fracture stress and strain and Young's modulus of flax yarn were measured

according to ASTM D 2256-97. Individual yarn specimens were tested on Instron testing machine, model 1122. A 100-mm length of yarn was used to determine the yarn diameter using an optical microscope, as described above. The same specimens were used for measuring their tensile properties. Tests were performed using a gauge length of 50 mm and at a strain rate of 1 min^{-1}. Twenty specimens were tested to obtain the average tensile properties.

2.7. Preparation of yarn reinforced composites

Unidirectional flax yarn reinforced composites were fabricated using MSPC-GP resin. Composites were prepared with yarns oriented in both longitudinal and transverse directions. Yarns were cut into 90 mm length and desired weight (nearly 1.85 g) was used for preparing each composite specimen. The yarn specimens had a high tendency to twist into a bundle, on contact with the resin, as a result of their high twist. As a result, extreme care was taken to align the yarns as parallel as possible. A rectangular, 15 × 70 × 15 mm, mold was prepared using Teflon® sheet for controlling the composite thickness. MSPC-GP resin was prepared as described earlier in Section 2.2. A layer of precured resin was poured over the mold and the parallel bundle of yarns was laid on the top. Since the yarns had very high twisting tendency, care was taken to keep the yarn bundle under tension all the time using a masking tape. Additional precured resin was poured very slowly on the top of the yarn bundle and another set of yarns was laid on the top. Finally, more resin was added on the top and the composite specimens were transferred into an air circulating oven maintained at 35°C for drying. The specimens were oven dried for nearly 48 h. These specimens were then transferred into a mold. A mold-release agent was used for easy removal of composites after curing. Curing was done in the Carver hydraulic hot press at 120°C for 25 min at a pressure of 8 MPa. On completion of the curing cycle, the specimens were removed from the hot press and conditioned at standard ASTM atmosphere prior to characterization of their properties. The fiber weight fraction in final composite specimen was calculated on the basis of final composite weight and initial weight of the yarn.

2.8. Tensile characterization of composites

Tensile properties of yarn reinforced composites were characterized in accordance with ASTM D 3039/D3039M-00. Three thickness measurements were made along the gauge length of each specimen and the average value was used for calculating the fracture stress and strain and Young's modulus. Specimens were approximately 1 mm thick and 10 mm wide. The tensile tests were performed on an Instron tensile tester, model 1122, at a strain rate of 1 min^{-1} and a gauge length of 50 mm. At least five specimens were tested to obtain the average tensile properties.

2.9. Flexural characterization of composites

Flexural properties of yarn reinforced composites were characterized in accordance with ASTM D 790-99. The specimens were approximately 1 mm thick and 10 mm wide. Three thickness measurements were made along the length of each specimen and the average value was used for calculating the flexural stress and flexural modulus. The flexural tests were carried out using an Instron tensile tester, model 1122 at a crosshead speed of 2 mm/min and span length of 20 mm. At least five specimens were tested to obtain the average flexural properties.

2.10. Fracture surface characterization of composites

The fracture surfaces of yarn reinforced composites were observed under Leica scanning electron microscope (SEM), model 440×. All fractured specimens were sputter coated with gold-palladium to get good conductivity. SEM photographs of specimens tested in both longitudinal and transverse directions were taken to characterize and understand the composite's failure mechanism. These photomicrographs were also used for qualitative analysis of interfacial bonding between the yarn and MSPC-GP resin. All SEM photographs were taken at the CCMR facilities.

3. Results and discussions

3.1. Effect of glycerin content on tensile properties

In part 1 of this paper we have shown that GA forms crosslinks with amine groups in SPC [10]. Since PVA contains hydroxyl groups and was expected to blend easily with SPC and GA and the MSPC-GP blend was expected to further increase the tensile properties by providing binding between SPC molecules. Based on the preliminary experiments, 40% GA with 40% PVA (w/w of SPC) was assessed to be the optimum blend composition for MSPC-GP resin preparation [11]. Glycerin content was varied between 0 and 15%, to study the effect of plasticizer content on the tensile properties of MSPC-GP resin. Adding PVA to SPC and GA blend, allowed to process films at very low content and even without any glycerin. However, only GA modified SPC resin could not be prepared with less than 10% glycerin content [10]. It is hypothesized that since low molecular weight PVA (M_w = 13,000–23,000) has been used in this research, it may act as a plasticizer. The films prepared with 0% glycerin showed no problem of curling or warping on the edges and could be easily processed.

Fig. 1 shows the effect of glycerin content on the tensile properties of MSPC-GP resin. As expected, increasing glycerin content from 0 to 15% increased the fracture strain from 1.8 to 17.5% and reduced the Young's modulus from 1.08 GPa to 412.6 MPa, respectively. The lower fracture stress values at very low glycerin concentration (0 and 5%) can be attributed to the brittle nature of MSPC-GP resin, leading to very low fracture strain values. As a result, the full fracture

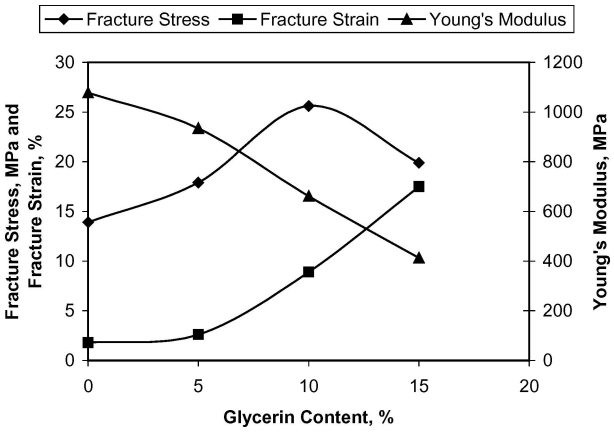

Figure 1 Effect of glycerin content on the tensile properties of MSPC-GP with 40% GA and 40% PVA.

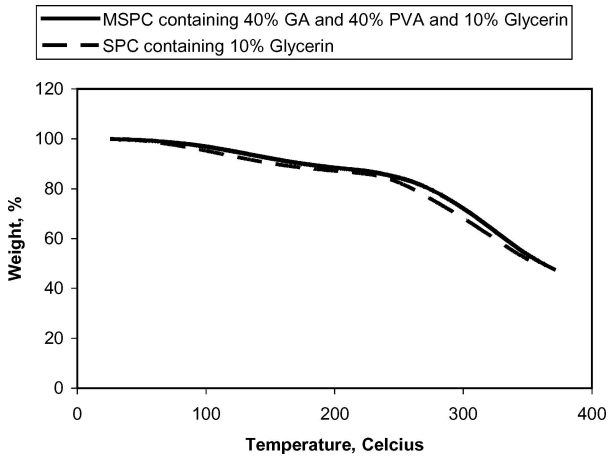

Figure 2 TGA thermogram of MSPC-GP resin containing 40% GA and 40% PVA and 10% glycerin.

stress value is not realized. As described earlier in part 1 of this paper glycerin acts as an effective plasticizer for SPC resin crosslinked using glutaraldehyde increasing its fracture strain significantly [10]. These results indicate that glycerin has a good plasticizing action with MSPC-GP, leading to reduced brittleness and increased plasticity. These results are in agreement with results reported by several other researchers [3, 7, 16–18].

3.2. Moisture absorption by SPC and MSPC-GP

Table I summarizes the effect of glycerin content on the moisture content of SPC and modified MSPC-GP resins. It is evident from Table I that reducing the amount of glycerin reduces the moisture content of both SPC and MSPC resins. All moisture content values were found to be statistically different at 95% confidence level and the standard deviations were found to be extremely low. As explained earlier in part 1 of this research [10], glycerin increases the free volume and decreases the glass transition temperature (T_g) of the system making it easier for the moisture to diffuse in. More importantly, due to the polar and hydrophilic nature of glycerin molecules resulting from the hydroxyl groups, water molecules are attracted to glycerin.

It can also be seen clearly that MSPC-GP resin has lower moisture content than SPC resin. This can be attributed mainly to the crosslinks formed by GA with amine groups of SPC [10]. Based on these results and ease of resin processing, MSPC-GP resin containing 40% GA and 40% PVA and 10% glycerin was selected as the optimum blend for composite fabrication. All results discussed henceforth are for this composition.

TABLE I Effect of glycerin content on the moisture content of SPC and MSPC-GP resins

Glycerin (%) (w/w of SPC)	Moisture content of SPC (%)	Moisture content of MSPC-GP (%)
0	N/A	11.3
5	13.2	11.8
10	13.7	12.3
15	15.0	13.1

3.3. Thermogravimetric properties of SPC and MSPC-GP

Fig. 2 shows a typical TGA thermogram for MSPC-GP resin, in nitrogen atmosphere. TGA thermogram of SPC resin has also been plotted in Fig. 2 for comparison. As can be seen from the thermograms, MSPC-GP resin shows improved thermal stability than SPC resin. MSPC-GP resin decomposes at a higher temperature (260°C) than SPC resin (235°C). The improved thermal stability of MSPC-GP resin is mainly attributed to the crosslinking between SPC molecules and GA. It can be also seen from the plot that the MSPC-GP resin is highly stable up to 120°C, the processing temperature for yarn reinforced composites. The weight loss up to 120°C is mainly due to the loss of moisture from the specimen.

3.4. Dynamic mechanical properties of SPC and MSPC-GP

Fig. 3 shows the plots of tan δ as a function of temperature for SPC resin containing 10% glycerin and MSPC-GP resin. It is evident from the plots that MSPC-GP resin shows the glass transition temperature (T_g) at 182°C and the β transition at 85°C, compared to a T_g of 173°C and the β transition at 80°C for SPC resin. The increase in T_g and β transition values of MSPC-GP

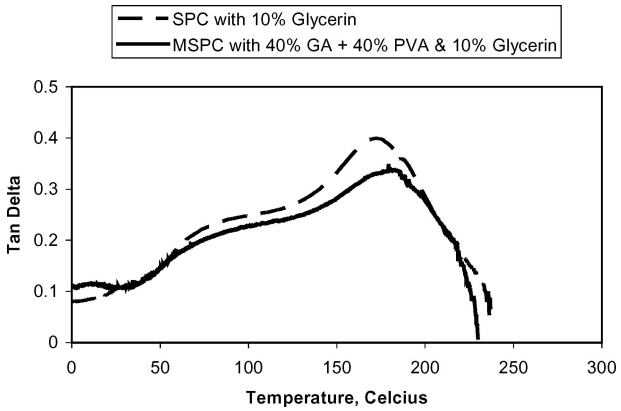

Figure 3 Variation in tan δ as a function of temperature for MSPC-GP resin.

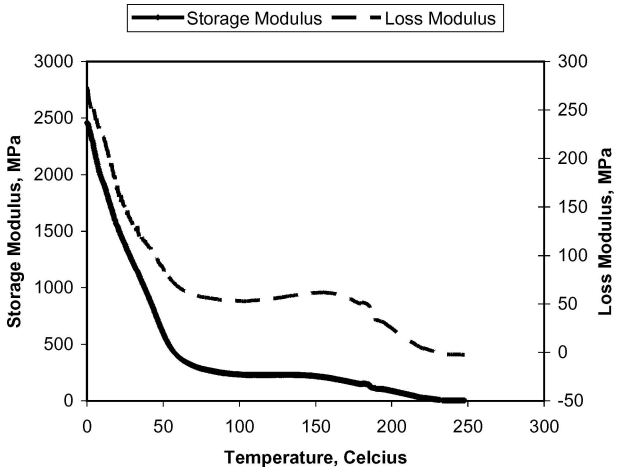

Figure 4 Variation in storage and loss moduli as a function of temperature for MSPC-GP resin.

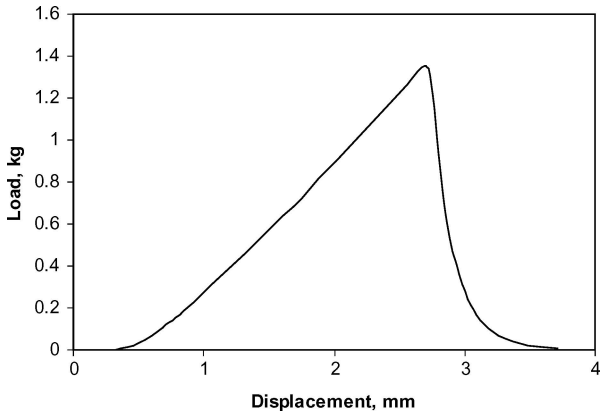

Figure 5 Typical load-displacement plot for flax yarn tensile test.

resin over SPC resin is attributed to the crosslinking of SPC with GA. However, the increase in T_g for MSPC-GP is 3°C lower than that obtained for GA crosslinked SPC (MSPC-G) resin (T_g = 185°C) [10]. This may be due to the plasticizing action of PVA in MSPC-GP resin.

Fig. 4 shows the storage modulus (E') and loss modulus (E'') as a function of temperature for MSPC-GP resin. The storage modulus value of MSPC-GP at 25°C is 1.4 GPa compared with 1.8 GPa obtained for SPC resin. The lower storage modulus for MSPC-GP resin is perhaps due to the plasticizing action of PVA molecules.

It can also be seen from Fig. 4 that the storage modulus of MSPC-GP resin decreases continuously with increase in temperature. However, there is no catastrophic change in E' value at T_g. This behavior is similar to results shown by other researchers [17, 18] and also by authors for SPC and GA modified SPC in part 1 [10].

3.5. Characterization of flax yarn

The flax yarns used in this study had an average diameter of 0.234 mm with a 14.7 percent coefficient of variation (CV%). The high CV% can be attributed to the large variation in the diameter of flax fibers and the spinning process. The high variation is seen in many other natural fiber yarns as well.

Tensile properties such as fracture stress, Young's modulus and fracture strain for flax yarn are summarized in Table II. As can be seen from Table II, there is a large variability in the fracture stress and Young's modulus values, which can be attributed to the variability in the flax yarn strength and also to the variation of yarn diameter along the length. Fig. 5 shows a typical load-displacement plot for the flax yarn. It can be seen from Fig. 5 that the flax yarns reach their highest load at about 4.9% strain and then fail catastrophically, indicating a brittle failure. This is attributed to the high twist yarn which does not allow any fiber slippage.

TABLE II Tensile properties of flax yarn

Fracture stress (MPa)	Fracture strain (%)	Young's modulus (GPa)
312 (24.5)*	4.9 (13.1)	8.5 (23.4)

*Figures in parentheses are CV%.

3.6. Tensile properties of composites

Tensile properties of unidirectional flax yarn reinforced MSPC-GP 'green' composites were characterized in both longitudinal and transverse directions. The composites had a fiber weight fraction of 45% and an average thickness of 1.1 mm. Table III summarizes the tensile properties of unidirectional yarn composites in both longitudinal and transverse directions. As can be seen from Table III, the composites have higher tensile properties in the longitudinal direction. This is due to the fact that the composite tensile properties are controlled by the yarn tensile properties in the longitudinal direction and by the resin tensile strength and/or interfacial properties in the transverse direction. The difference between the composite tensile properties in the longitudinal and transverse directions was tested for significance and was found to be significant at 95% confidence level, as expected.

Fig. 6 shows typical load displacement plots for the flax yarn reinforced MSPC-GP composites in both longitudinal and transverse directions. Fig. 7 shows a typical tensile fracture sequence of a flax yarn reinforced MSPC-GP composite in the longitudinal direction. As can be seen from Figs 6 and 7, the composite specimens attain their highest load and then fail catastrophically when tested in the longitudinal direction. The brittle fracture behavior of the composites is similar to the fracture of flax yarns, as shown in Fig. 5. It can also

TABLE III Tensile properties of flax yarn reinforced MSPC-GP composites

Test direction	Fracture stress (MPa)	Fracture strain (%)	Young's modulus (GPa)
Longitudinal	126 (13.6)*	11.6 (7.6)	2.24 (8.8)
Transverse	3.9 (36.8)	1.5 (36.0)	0.83 (30.7)

*Figures in parentheses are CV%.

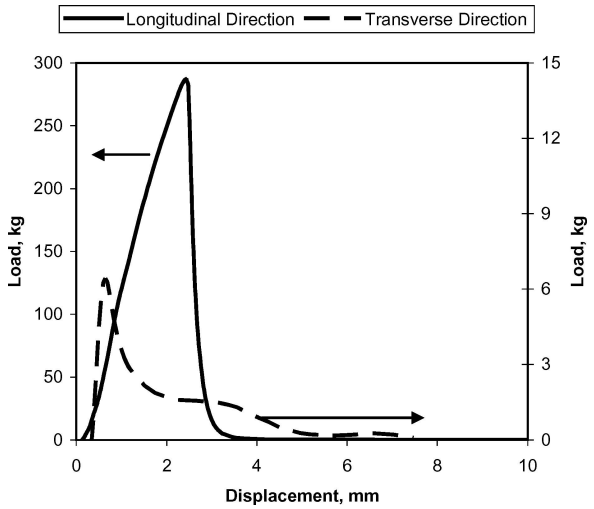

Figure 6 Typical load vs displacement plots for flax yarn reinforced MSPC-GP composites during tensile tests.

be seen from Fig. 6 that in the transverse direction, the composite specimens do not show catastrophic failure. As mentioned earlier, most short staple yarns such as the flax yarns used in this study have hairiness i.e., short fibers protruding out. These fibers either get entangled with the neighboring yarns or embed into the resin. In addition to the yarn hairiness, non-aligned fibers resulting from resin shrinkage (explained in the following paragraphs) as well as twisting of the yarns, though controlled to some extent during composite fabrication, help prevent catastrophic failure, as can be seen in Fig. 6.

Mohanty *et al*. [12] have investigated the effect of various chemical modifications of the jute yarn on the mechanical properties of biodegradable jute yarn-Biopol^{TM} composites. They showed that the alkali treated jute yarn reinforced composites, with approximately 23% fiber weight fraction, increased the composite tensile strength by 294% as compared to the pure resin. In the present research, flax yarn reinforced composites increased the tensile strength by 493% (126 MPa) as compared to the MSPC-GP resin (25.6 MPa). This improvement is similar since the composites prepared in this study had 45% weight fraction of flax yarns. Good bonding between the MSPC-GP resin and the strength of the flax yarn contribute to the high tensile properties.

Simple rule of mixture [19] was used for predicting the longitudinal fracture stress and Young's modulus for flax yarn reinforced MSPC-GP composites. The

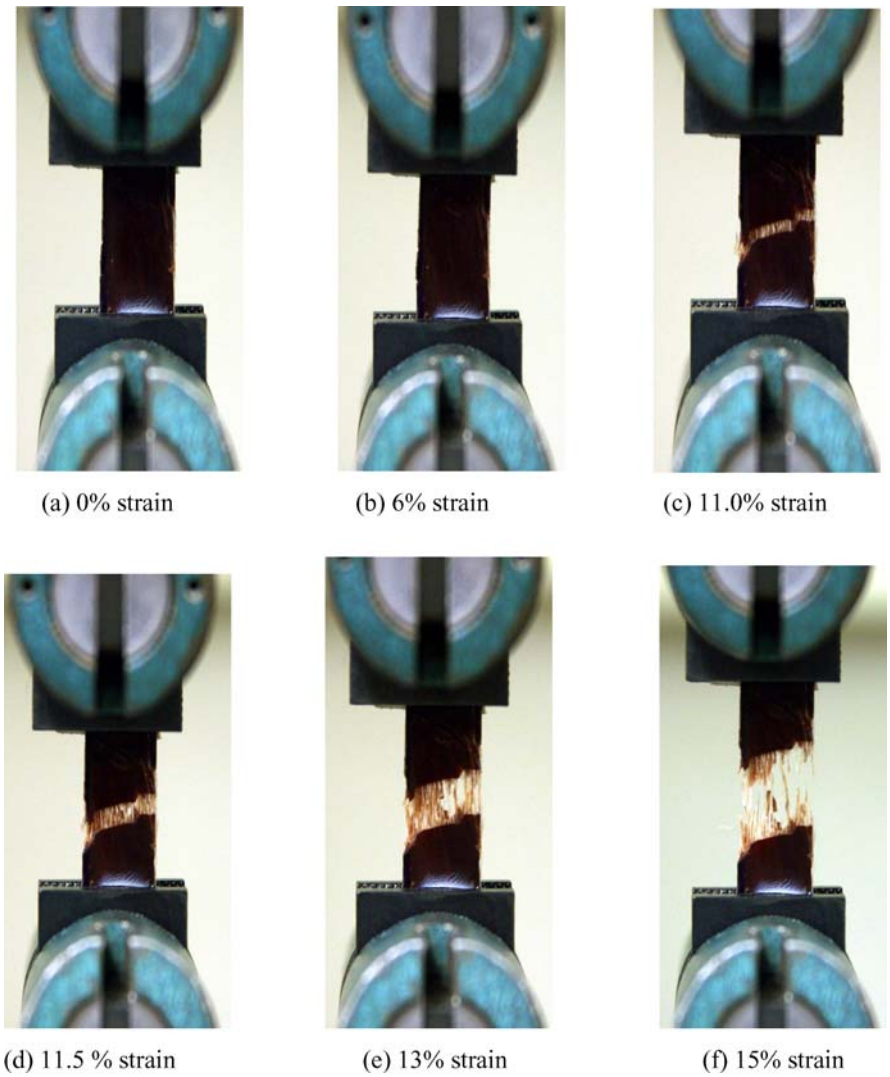

Figure 7 Photographs showing typical tensile fracture of flax yarn reinforced MSPC-GP composites in the longitudinal direction.

rule of mixture evaluates the fracture stress and Young's modulus values as shown below in Equations 1 and 2, respectively:

$$\sigma_c = \sigma_y V_y + \sigma_m V_m \quad (1)$$

$$E_c = E_y V_y + E_m V_m \quad (2)$$

where σ_c, σ_y and σ_m are the fracture stress of the composite, yarn and matrix, respectively. E_c, E_y and E_m are the Young's modulus for the composite, yarn and the matrix, respectively. V_f and V_m are the volume fractions of the fiber and the matrix, respectively. The fracture stress and Young's modulus values for composites calculated using the rule of mixture were 145 MPa and 3.9 GPa, respectively whereas the experimental values for the composite fracture stress and Young's modulus were 126 MPa and 2.24 GPa, respectively. One of the primary reasons the experimental values for both fracture stress and Young's modulus are lower than the theoretical predictions is the resin shrinkage during curing. As described earlier in Section 2.2, water was used, in a ratio of 15:1 (w/w of SPC), during precuring of MSPC-GP resin. The loss of all this water during drying resulted in significant shrinkage of the resin. In the present study the yarns were not held taut, under tension, during the drying and curing of the composites. Resin shrinkage caused the yarns to undergo longitudinal compression, which was overcome during tensile testing. This is believed to be the major factor reducing the composite modulus [7]. The difference between the theoretical and experimental moduli could be reduced by keeping yarns under tension and thus avoiding their shrinkage. Although care was taken to align the yarns parallel to each other, the yarns showed a tendency to twist into a bundle on contacting MSPC-GP resin as explained earlier in Section 2.7. All these deviations from the perfect parallel alignment contributed towards lower experimental modulus values. It is important to note that the resin shrinkage, to some extent, is also responsible for the yarn misalignment. Some voids present in the composites also lead to lower experimental values. Voids are inevitably generated in the composite due to water vaporization in MSPC-GP resin and flax yarn during drying and especially during curing at 120°C. Similar void problems have been reported by Nam [7] and Lodha and Netravali [3], while fabricating unidirectional ramie fiber/SPC resin and short ramie fiber/SPI resin composites, respectively.

3.7. Flexural properties of composites

Table IV summarizes the flexural properties including flexural stress and strain and flexural modulus of flax yarn reinforced composites, at yield. As can be seen from Table IV, the composite specimens have higher flexural properties in the longitudinal direction. The difference between the composite tensile properties in the longitudinal and transverse directions was tested and found to be significant at 95% confidence level. Once again these results show that the flax yarn properties control the flexural properties of the composite in the longitudinal direction, while the MSPC-GP resin properties control the flexural properties in the transverse direction as would be expected. The reasons for these differences are same as explained in the Section 3.6 for composite tensile properties.

TABLE IV Flexural properties of flax yarn reinforced MSPC-GP composites

Test direction	Flexural stress (MPa)	Flexural strain (%)	Flexural modulus (GPa)
Longitudinal	86 (13.2)*	15.1 (17.5)	1.18 (23.8)
Transverse	3.6 (27.4)	8.15 (24.0)	0.055 (36.5)

*Figures in parentheses are CV%.

Nam [7] fabricated unidirectional composites using ramie fiber and SPC resin. She reported the flexural stress and flexural modulus values of 225 MPa and 12.3 GPa, respectively, in the longitudinal direction. The lower flexural stress and modulus values obtained

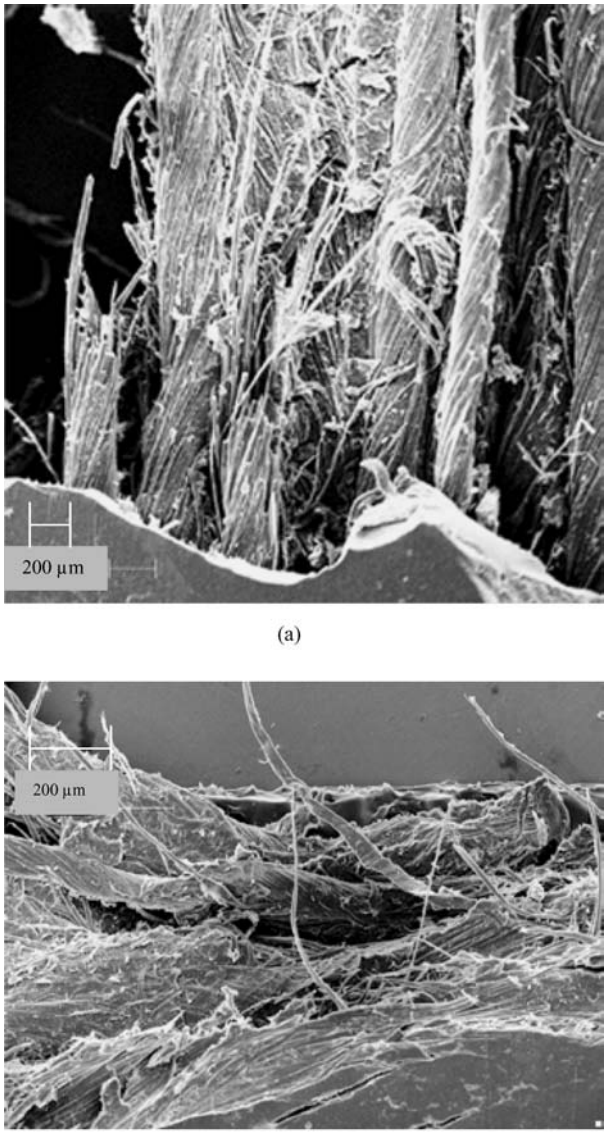

Figure 8 SEM photographs of the fracture surfaces of flax yarn reinforced MSPC-GP composites in the tensile mode in (a) longitudinal and (b) transverse directions.

in this research are due to the yarns used in this research which have lower strength and modulus as compared to ramie fibers used by Nam [7].

3.8. Fracture surface characterization of composites

Figs 8a and b show typical SEM photographs of the tensile fracture surfaces of the flax yarn reinforced MSPC-GP composites in the longitudinal and transverse directions, respectively. It can be seen from Figs 7 and 8a that the resin is adhering to the yarn surface, after the fracture, indicating good bonding between the MSPC-GP resin and the flax yarn. As explained earlier, this is due to the polar nature of the flax (cellulosic) fiber and the polar groups present in soy protein resin. It can be confirmed from Fig. 6 that the composites undergo a brittle failure, reflecting the brittle fracture of flax yarn.

Fig. 8b shows the SEM photograph of a typical tensile fracture surface of the flax yarn reinforced MSPC-GP composite in the transverse direction. It can be seen from Fig. 8b that some of the fibers protrude out of the fracture surface. These fibers bridge the resin and are responsible for continuing to carry a fraction of the load after the resin failure. Also, as explained earlier in Section 3.6, yarn hairiness and non-alignment also contribute towards a non-catastrophic failure. As a result, the load doesn't drop to zero immediately. This behavior is clearly seen in the load vs. displacement plot, in transverse direction, shown in Fig. 6. It can be seen in the fracture surface shown in Fig. 8b where the fibers and yarns are seen splitting outwards with resin attached on their surface.

4. Conclusions

SPC was blended with PVA and cross-linked using GA. The blend allowed to process SPC resin without any plasticizer e.g. glycerin. The modified resin showed improved tensile properties such as fracture stress and Young's modulus. MSPC-GP resin also showed improved thermal stability and increased the glass transition temperature. The MSPC-GP resin also exhibited reduced moisture content than the SPC resin. Flax yarn and MSPC-GP resin were used to fabricate fully biodegradable, environment friendly 'green' composites. Flax yarn reinforced unidirectional MSPC-GP composites, with 45% fiber weight fraction, exhibited significantly higher fracture stress and Young's modulus in the longitudinal direction as compared to the MSPC-GP resin. The properties in the transverse direction are controlled by the resin or interface properties. As a result, the strength and modulus in the transverse directions were significantly lower than those obtained in the longitudinal direction. The theoretical analysis for composite fracture stress and Young's modulus in the longitudinal direction predicted higher values than the experimental results. This deviation is mainly a result of longitudinal yarn compression during processing as a result of resin shrinkage. Less than perfect yarn orientation as well as voids in the composites, which could be improved by improving processing, are also factors contributing to the lower mechanical properties. These 'green' composites confirmed good bonding between flax yarn and MSPC-GP resin, which is expected because of their polar chemistry. However, composite strength and bonding can be further improved by various chemical modifications of MSPC-GP resin and surface modifications of the flax yarn as well as better processing techniques.

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