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ScienceDirect
Procedia CIRP 17 (2014) 778 – 782
www.elsevier.com/locate/procedia

Variety Management in Manufacturing. Proceedings of the 47th CIRP Conference on Manufacturing Systems

A novel process for the production of unidirectional hybrid flax/paper reinforcement for eco-composite materials

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Abstract

In this paper a new process to manufacture unidirectional reinforcements for eco-composite materials, made of natural fibers, is presented. Starting with flax rovings of different sizes, an apparatus was developed to feed and align the rovings over the wet-end section of a paper machine. The short kraft paper fibers are therefore mixed with the long flax roving as the machine is running, and at the end of the process, a sheet of the hybrid dry reinforcement is obtained and cut to size for impregnation with various resins, using different processes. This novel manufacturing process allows for high volume production of reinforcement. It is very flexible, and many different combinations of long and short fibers can be exploited for the production of a vast variety of dry reinforcements. In this paper, composite samples are obtained out of these reinforcements, using the resin infusion (RI) molding process with a commercial epoxy resin. The results are compared with those of usual glass fiber reinforcement. An interesting aspect is that the large variability, typical for natural fibers, is largely reduced when the short kraft fibers are present in the composite. In terms of permeability to resin, reasonably comparable values can be obtained compared to that of glass fabrics, if a low surface density of reinforcement is chosen.

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Keywords: Paper machine; long flax roving; unidirectional hybrid reinforcement

1. Introduction

These days the pulp and paper industry is experiencing difficulties due to the increasing tendency toward the use of electronic media resulting in a lower use of hardcopies. On the other hand, in terms of imparting flax fibers as reinforcing agent in composite materials, short fibers in the form of randomly oriented reinforcements (mat) have been already implemented to produce non-structural parts such as car interior trims [1]. Due to the higher price of developing unidirectional or woven natural fiber fabrics, few studies have dealt with developing continuous natural fiber reinforcements for load bearing application composite parts.

Goutianos et al. [2] produced some textile reinforcements including uniaxial and biaxial warp knitted fabrics as well as biaxial plain weaves, based on flax yarns. The mechanical properties of composite materials fabricated out of these fabrics indicate that, although the strength is inferior to that of woven glass composites, the stiffness and particularly the specific stiffness (stiffness divided by density) can compete with woven glass composites. Pothan et al. [3] in 2008, developed three different sisal fiber based textiles, namely plain, twill and matt weave architecture. The weaving pattern with maximum fibers in the loading direction (matt architecture) is reported the best one to improve composite properties. In 2011, Miao and Shan [4] modified the conventional nonwoven mat manufacturing process and produced highly aligned nonwoven mats of flax fibers as an alternative to unidirectional fabrics. It is claimed that, while the reinforcement production cost is more budget than unidirectional flax/PP fabric, the composites show similar strength, although elastic modulus is lower. Recently, in 2013

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doi:10.1016/j.procir.2014.01.108

Xue and Hu [5] developed a biaxial weft-knitted flax fabric using a modified flat knitting machine. The modulus and the strength of the resulting composites are reported 4.2GPa and 161.6MPa, respectively, along the weft direction. . Similarly Muralidhar et al. [6, 7] made woven and rib-knitted flax reinforcement and characterized their composite material.

While the majority of research works on continuous plant fiber reinforcements are devoted to developing bi-axial fabrics using textile industry machinery, it is a novel idea to produce unidirectional flax fiber reinforcement with papermaking machines. Considering the high volume production capacity of these machines combined to more than a century of machine developments for the production of high quality papers, these equipments represent an interesting research and development avenue to produce high quality natural fiber reinforcements. A new potential market to pulp and paper industry could be developed by producing high quality and low cost reinforcements for the composite industry, using the paper phase as a thin binder layer for the long natural reinforcing fibers. On top of that, this represents an opportunity to produce added-value unidirectional natural fiber reinforcements with budget price, due to mass scale production, inducing at the same time superior mechanical properties to composite materials owing to the unidirectional architecture of the reinforcement. Considering the increasing demand for the use of bio-compatible materials due to the global ecological awareness, the new materials could well respond to this tendency.

2. Basic papermaking process

The paper making process starts with wood chips as the chief raw material and stops with the paper sheet coming off the paper machine. As is schematically outlined in the fig 1 the wood chips process through the pulp mill, the stock (furnish) preparation facilities and the paper machine, respectively.

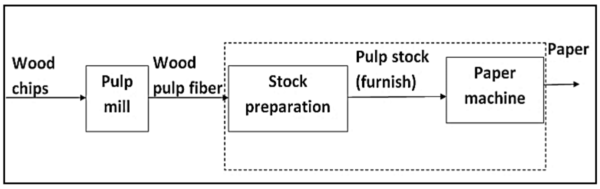

Fig. 1. Schematic of the paper processing

The wood chips used for paper production comes either from hardwood (deciduous) trees like polar or eucalyptus or softwood (conifer) trees such as fir and pine. Softwood has longer fiber than deciduous species and generally forms stronger papers. In this study softwood kraft pulp provided by Innofibre[1] is employed.

In the pulp mill, the fibers are separated from wood chips while, in the next step of stock preparation, a refined pulp slurry is prepared for the paper machine. Modern papermaking machines, which are named after the first Fourdrinier brothers' paper machine back in the 1804 [8], include five major divisions, namely, the headbox, the forming table, the press section, the dryer section and the calendaring section. While the first three segments are referred to as "wet end" the last two divisions are named "dry-end". These divisions are schematically illustrated in Fig. 2.

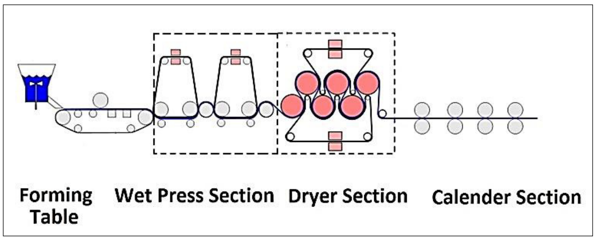

Fig. 2. Schematic of a Fourdrinier paper machine
(www.wikipedia.com, Author: Egmason)

The experimentation in the present study is performed on the Fourdrinier pilot-scale paper machine of Innofibre. The specifications of this machine are given in table 1. The main difference between a pilot paper machine and an industrial one is the width of the forming table, which is up to several meters in the case of an industrial machine.

Table 1. General characteristics of the paper machine employed for this study.

Forming table	Maximum width (m)	0.45
	Table length(m)	21
Presses	1st step(kN/m)	0-100
	2nd step (kN/m)	0-350
	3rd step (kN/m)	0-1000
Dryers	Total number of cylinders	28

Once the pulp slurry of very low consistency (solid content) is spread evenly over the forming table, using the headbox, the dewatering starts through gravity force and the solution reaches 20% consistency at the end of the forming table. Similarly, at the end of the press section, some 15-25% of water is squeezed out and the sheet reaches up to 35-45% of solid content. At this stage, there is still a considerable amount of water in the paper. So, in the drying section the majority of water content is vaporized and at the end of the drying section, the paper sheet reaches 94-97% of consistency.

3. Feeding the flax roving

In order to develop the hybrid-reinforcement, flax roving must be introduced into the paper machine while it is running. This is accomplished using the frame shown in fig. 3, which is particularly designed for this purpose. The frame can accommodate up to 16 bobbins over which the desired lengths of roving are wound. The roving ends are then passed through

[1] - www.innofibre.ca

the bars containing holes (eyelet bars) and subsequently through the tightener section. Next, the spacing between the rovings is adjusted using calibrated combs to the desired pattern. Up to now, three combs have been designed but only two of them are tested.

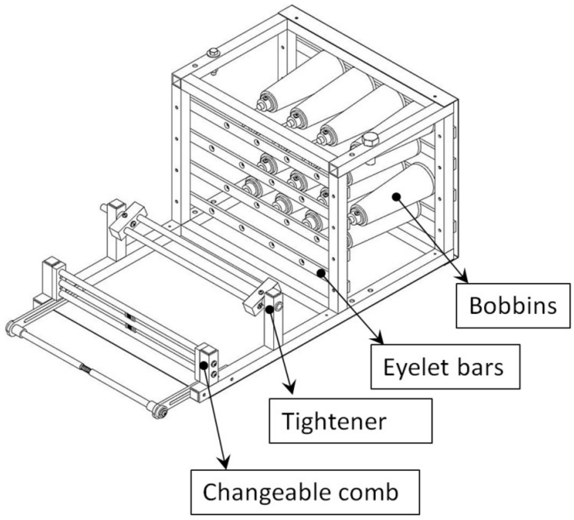

Fig. 3. Frame to feed and deposit flax roving into paper machine

After sufficient length of roving is wound around the bobbins and the roving ends are passed through the eyelet bars, the tightener and finally the comb, the feeding frame is installed on the paper machine. Fig. 4 shows the feeding frame installed over the forming table. This frame can be positioned to the desired location along the machine and at the desired angle with respect to the top of the table.

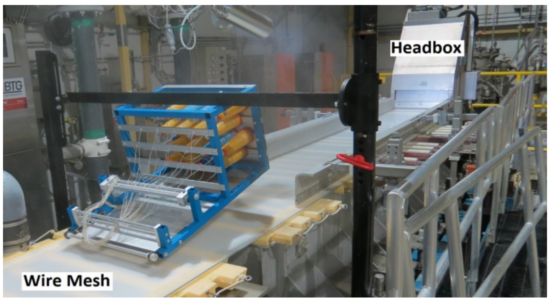

Fig. 4. Installed feeding frame on the paper machine table

When the ends of the flax roving contacts the forming table over which the pulp slurry is already spread (using the headbox), the hydrodynamic forces cause the flax roving to be drawn into the paper machine. At the end of the machine, unidirectional hybrid flax/paper reinforcement emerges, to be used later for the molding of multilayer composite parts. Figs. 5 and 6 reveal two very different reinforcement samples that were produced. The surface density of paper in these experimentations is 90 g/m^2 and a tex 200 flax yarn (200 grams per 1000 meters of yarn length), supplied by Safilin Inc. (Szczytno, Poland), is employed.

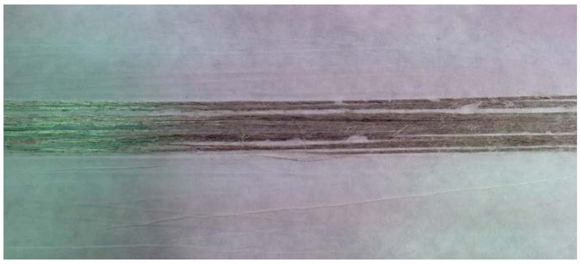

Fig. 5. Reinforcement sample produced with 16 roving in one inch

As can be noted, a large variety of dry reinforcement can be produced simply by adjusting the frame in figure 3. The reinforcement produced in figure 5 could be used for structural applications where the yarns are deposited side-by-side to produce a high stiffness and strength composite layer. So, many layers at different orientations, to suit specific needs, would make up the final product. The reinforcement in figure 6 could be used for packaging, where the flax rovings provide higher strength than paper or cardboard alone. In both cases different combinations of pulps, chemical additives and impregnation resins can be used, yielding an almost infinite number of combinations. Our studies aim exactly at understanding the functional relationship between the raw material properties and manufacturing process parameters, considered as inputs, and the final eco-composite properties measured after impregnation and resin cure, as outputs.

Fig. 6. Reinforcement sample produced with 10 roving, one inch spacing between each

4. Properties of the resulting eco-composites

An alternative way to the mass scale fabrication of the hybrid reinforcement on the paper machine is laboratory-scale fabrication. Using laboratory-scale fabrication set-up one can develop the reinforcements more rapidly and at a lower cost. Then, based on the analysis, get insight into the behavior of the process and materials for the scale-up to the real paper machine.

Fig. 7 illustrates the laboratory-scale fabrication process. It consists of four main steps namely preparing pulp slurry (step 1), fabrication of paper sheet with a dynamic sheet former (step 2), introducing a pre-aligned flax ply upon the paper sheet through pressing (step 3), and finally drying the hybrid-reinforcement (step 4). The second, third and fourth steps of the laboratory-scale reproduction are the respective counterparts of the forming table, press and dryer sections of the paper machine (shown in fig 2). A typical sample fabricated in the laboratory is shown in Fig. 8. The reinforcements used for the purpose of testing in this study, are prepared in the laboratory, using on the above-mentioned procedure.

Fig. 7. Processing of the reinforcement in laboratory

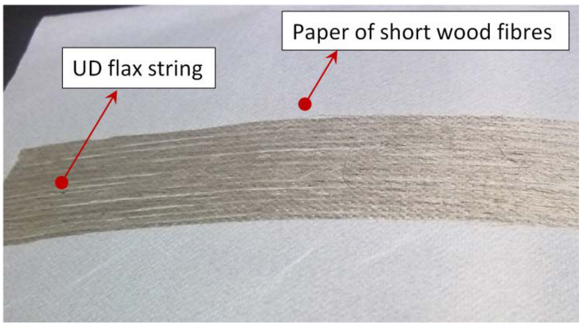

Fig. 8. A typical reinforcement sample made in the lab

Composite plaques are fabricated by the hand-layup method using a low viscosity Adtech™ 820 epoxy laminating resin. The samples were cured overnight in a vacuum bag under 20 in-Hg (67.7 kPa) pressure and finally undergone a post cure at 75°C for 16h, before testing. At least five tensile test coupons were cut from the plaques for testing.

Fig. 9 shows typical fractured coupons for two eco-composite samples. In coupon (A), the hybrid flax/paper reinforcement is used while in coupon (B), the paper layer was excluded from the reinforcement. As can be clearly seen, a more uniform confined fracture surface is observed for coupon (A). Such behavior was systematically reproduced for all coupons. Furthermore, the tensile strength of sample (A) was about 25 MPa higher than sample (B), although some marginal reduction was also observed in elasticity modulus of the specimen (A). Most importantly, the variability (standard deviation) of the tensile strength results was reduced approximately by half in the case of flax/paper composites compared to flax composites. These results suggest that a more repetitive behavior can be obtained for the hybrid unidirectional flax/paper reinforcement compared to the unidirectional flax only composites and that, flax/paper composite is less sensitive to the sources of variability in natural fibers.

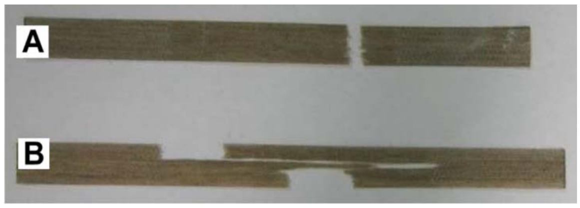

Fig. 9. Composite samples with (A) hybrid unidirectional flax/paper reinforcement (B) unidirectional flax ply alone

Table 2- measured properties of eco-composite materials

Property	UD flax/paper/epoxy[9]	UD glass/epoxy[10]
paper surface density (g/m^2)	88	-
flax surface density (g/m^2)	180 (roving of tex 200)	-
V_f (%)	46.4	33.5
strength(MPa)	173	549
stiffness (GPa)	12.720	24.2
specific strength (MPa/gr.cm^{-3})	115.33	214.45
specific stiffness (GPa/gr.cm^{-3})	8.48	9.45

Table 2 compares the tensile test results of samples obtained with the flax/paper reinforcement with those of glass fiber reinforced epoxy obtained from the literature [10]. For the flax/paper/epoxy composite, a specific elasticity modulus (elasticity modulus divide by the fiber density) of 8.48 GPa/gr.cm^{-3} and a specific tensile strength of 115.33 MPa/gr.cm^{-3} are observed, for a fiber volume fraction of 46.4%. Corresponding values of 9.45 GPa/gr.cm^{-3} and 214.45 MPa/gr.cm^{-3} were respectively reported for the glass fiber composite, at a V_f of 33.5%. To compare the results on the base of similar fiber contents, the flax/paper/epoxy results can be multiplied by the fiber content ratios. This can be done if we consider that the rule of mixture, commonly used for unidirectional composites, is proportional to the fiber content. A specific stiffness of 33.5/46.4*8.48=6.12 GPa /gr.cm^{-3} and a specific strength of 33.5/46.4*115.33= 83.26 MPa/gr.cm^{-3} are thus obtained for flax/paper/epoxy composite at 33.5% V_f. Comparing the above-calculated values with UD glass/epoxy (table 2) it can be noticed that the specific elasticity modulus is quite close to that of unidirectional glass fiber composite while the specific strength has been reduced by 60%.

Results of permeability measurement are shown in table 3, where the measurements are performed based on ref. [11]. From table 3, it turns out that the surface density of flax layer plays an important role in deciding the permeability magnitude. For a surface density of about 426 g/m^2 (second column of table3), the observed permeability is extremely low and of no practical interest. However, when the flax surface density is reduced to 162.8 g/m^2 (fourth column of table3), while paper surface density remains almost the same (around 40 g/m^2), permeability along the fiber direction (K_x) rises to

22.2×10^{-12} m^2. If the paper surface density is further reduced to about 28.9 g/m^2 (third column of table3) the along-fiber permeability will be approximately doubled and becomes 42.7 $\times10^{-12}$ m^2. In addition, the cross-fiber permeability (K_y) raises to 8 $\times10^{-12}$ m^2. Corresponding values for a woven cross-ply glass fabric (fifth column of table3) are 109.53e^{-12} m^2 and 73.5e^{-12} m^2, respectively. While K_x of the latter hybrid reinforcement is about 40% of glass fabric, K_y is much lower, with a value around just 10% of glass fabric.

Table 3- outcomes of permeability test

Property	UD flax/Paper[9]	UD flax/Paper	UD flax/Paper	Woven glass [9]
Paper surface density (g/m^2)	40	28.9	38.8	805
Flax surface density (g/m^2)	426 (roving of tex 1000)	128.7 (roving of tex 200)	162.8 (roving of tex 200)	
Nb of stacked layers	4	3	3	3
V_f (%)	41.1	35.0	35.0	41.1
K_{xx} (m^2 $\times 10^{-12}$)	very low	42.7	22.2	109.53
K_{yy} (m^2 $\times 10^{-12}$)	Very low	8.0	3.26	73.5

5. conclusion

A novel bio-based reinforcement consisting of a unidirectional flax roving layer deposited over a tiny layer of kraft paper was successfully manufactured using a pilot-scale paper machine. To feed the flax into the paper machine a frame accommodating up to 16 bobbins was installed over the forming table of paper machine at the wet-end section. Employing two different combs to adjust the flax roving spacing, two types of reinforcements was produced, one with 16 roving per inch and another with one roving per inch which could be used to produce reinforced paper only.

Composite samples were fabricated out of the hybrid unidirectional flax/paper reinforcement and their tensile behavior is compared with unidirectional flax fiber reinforced epoxy. The comparison shows that at the present of paper layer, the variance in the composite properties is reduced. Comparing the tensile results also reveals that specific stiffness is reasonably comparable with that of unidirectional glass fiber composites.

Permeability results indicate the noticeable influence of flax layer surface density. For the lowest value of reinforcement surface density which results in the highest permeable preform, the acquired permeability accounts for 40% of woven glass fabric permeability.

Globally, the results are promising, but more development is required to get optimized results through tailoring both the process and material parameters. Some of the parameters to be adjusted include pressure and temperature of the fabrication process, the surface density of flax and paper layers, the yarn's tex value, their spacing, the type of pulp, etc. The novel process is therefore very flexible and it is believed that a large variety of custom end-product properties can be obtained using the same process.

Acknowledgements

The authors would like to acknowledge the financial support of Natural Sciences and Engineering Research Council (NSERC) of Canada. The authors also acknowledge the collaboration of the Innofibre research group of CEGEP of Trois-Rivières for production of the reinforcement on their pilot paper machine.

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